

Encyclopedia of Materials:
COMPOSITES

Editor in Chief
Dermot Brabazon

Section Editors

Manoj Gupta

Fatima Zivic

Eva Pellicer

Robertt Valente

Mohamed El Mansori

Lorna Fitzsimons

Antonello Astarita



ENCYCLOPEDIA OF MATERIALS: COMPOSITES

Volume 3

ENCYCLOPEDIA OF MATERIALS: COMPOSITES

EDITOR IN CHIEF

Dermot Brabazon

*I-Form, Advanced Manufacturing Research Centre, and Advanced Processing Technology
Research Centre, School of Mechanical and Manufacturing Engineering,
Dublin City University, Dublin, Ireland*

Volume 3

Section Editors

Section 6: *Design Methods for Composite Materials*, Edited by Robertt Valente

Section 7: *Nature Based and Inspired Composite Materials*,

Edited by Mohamed El Mansori

Section 8: *Life Cycle Analysis and Sustainability of Composite Materials*,

Edited by Lorna Fitzsimons

Section 9: *Joining of Composite Materials*, Edited by Antonello Astarita



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LIST OF CONTRIBUTORS FOR VOLUME 3

Faris M. Al-Oqla
The Hashemite University, Zarqa, Jordan

Carlos R. Alcalá
Airbus Operations SL, Cadiz, Spain

Dragan Aleksendrić
Faculty of Mechanical Engineering of the University of Belgrade, Belgrade, Serbia

Amit Arora
Advanced Materials Processing Research Group, Indian Institute of Technology Gandhinagar, Gandhinagar, Gujarat, India

Antonello Astarita
Department of Chemical, Materials and Industrial Production Engineering, University of Naples Federico II, Naples, Italy

Christophe Baley
South Brittany University, Lorient, France

Mariana D. Banea
Federal Center of Technological Education of Rio de Janeiro, Rio de Janeiro, Brazil

A.J. Baptista
Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal

Alberto Barroso
Sevilla University, Seville, Spain

Moisés Batista
University of Cadiz, Cadiz, Spain

Jorge Belinha
Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal and Polytechnic of Porto, Porto, Portugal

Eva Binder
Tongji University, Shanghai, China; TU Wien, Vienna, Austria; and Linnaeus University, Växjö, Sweden

Antonio Blázquez
Sevilla University, Seville, Spain

Alain Bourmaud
South Brittany University, Lorient, France

Dermot Brabazon
I-Form, Advanced Manufacturing Research Centre, and Advanced Processing Technology Research Centre, School of Mechanical and Manufacturing Engineering, Dublin City University, Dublin, Ireland

Raul D.S.G. Campilho
Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal and Polytechnic of Porto, Porto, Portugal

Ricardo J.C. Carbas
University of Porto, Porto, Portugal

Pierpaolo Carlone
University of Salerno, Fisciano, Italy

Felipe Cerdas
Chair of Sustainable Manufacturing and Life Cycle Engineering, Institute of Machine Tools and Production Technology (IWF), Technische Universität Braunschweig, Braunschweig, Germany

Faissal Chegdani
Arts et Metiers Institute of Technology, Mechanics Surfaces and Materials Processing, HESAM University, Châlons-en-Champagne, France

Lucas F.M. da Silva
University of Porto, Porto, Portugal

Irene Del Sol
University of Cadiz, Cadiz, Spain

Roberta Della Gatta
Department of Chemical, Materials and Industrial Production Engineering, University of Naples Federico II, Naples, Italy

Ivan Deviatkin
Department of Sustainability Science, Lappeenranta-Lahti University of Technology LUT, Lappeenranta, Finland

Lúcia M.J.S. Dinis
Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal and Faculty of Engineering of the University of Porto, Porto, Portugal

Jasmin Dönmez
Chair of Sustainable Manufacturing and Life Cycle Engineering, Institute of Machine Tools and Production Technology (IWF), Technische Universität Braunschweig, Braunschweig, Germany

Mohamed El Mansori
Arts et Metiers Institute of Technology, Mechanics Surfaces and Materials Processing, HESAM Université, Châlons-en-Champagne, France and TX A&M Engineering Experiment Station, Institute for Manufacturing Systems, College Station, TX, United States

Severo R. Fernandez-Vidal
University of Cadiz, Cadiz, Spain

Lorna Fitzsimons
Advanced Processing Technology Research Centre, School of Mechanical and Manufacturing Engineering and the Water Institute, Dublin City University, Dublin, Ireland

Antonio J. Gamez
University of Cadiz, Cadiz, Spain

Piyush Gohil
The Maharaja Sayajirao University of Baroda, Baroda, Gujarat, India

Alvaro Gomez-Parra
University of Cadiz, Cadiz, Spain

Vannessa Goodship
University of Warwick, Coventry, United Kingdom

Kaisa Grönman
Department of Sustainability Science, Lappeenranta–Lahti University of Technology LUT, Lappeenranta, Finland

Anurag Gumaste
Advanced Materials Processing Research Group, Indian Institute of Technology Gandhinagar, Gandhinagar, Gujarat, India

Pedro J. Herrera-Franco
Marista University of Mérida, Mérida, Yucatán, México and Scientific Research Center of Yucatan, Mérida, Yucatán, México

Christoph Herrmann
Chair of Sustainable Manufacturing and Life Cycle Engineering, Institute of Machine Tools and Production Technology (IWF), Technische Universität Braunschweig, Braunschweig, Germany

Mohamad R. Ishak
Universiti Putra Malaysia, Serdang, Selangor, Malaysia

Mohammad Jawaid
Universiti Putra Malaysia, Serdang, Selangor, Malaysia

Maya J. John
Centre for Nanostructures and Advanced Materials, Council for Scientific and Industrial Research, Pretoria, South Africa; Department of Chemistry, Nelson Mandela University, Port Elizabeth, South Africa; and School of Mechanical, Industrial and Aeronautical Engineering, University of the Witwatersrand, Johannesburg, South Africa

Renato M.N. Jorge
Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal and Faculty of Engineering of the University of Porto, Porto, Portugal

Jesús Justo
Sevilla University, Seville, Spain

Alexander Kaluza
Chair of Sustainable Manufacturing and Life Cycle Engineering, Institute of Machine Tools and Production Technology (IWF), Technische Universität Braunschweig, Braunschweig, Germany

Eric Le Bourhis
Institut Pprime, CNRS-Université de Poitiers-ISAE-ENSMA UPR, Département Physique et Mécanique des Matériaux, SP2MI, bd M&P Curie, Futuroscope, France

Zulkiflle Leman
Universiti Putra Malaysia, Serdang, Selangor, Malaysia

Xian Liu
Tongji University, Shanghai, China

Valentina Lopresto
Department of Chemical, Materials and Production Engineering, University of Naples Federico II, Naples, Italy

V.P. Mahesh
Advanced Materials Processing Research Group, Indian Institute of Technology Gandhinagar, Gandhinagar, Gujarat, India

Herbert Mang
Tongji University, Shanghai, China and TU Wien, Vienna, Austria

A.T. Marques
Faculty of Engineering of the University of Porto, Porto, Portugal and Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal

Eduardo A.S. Marques
University of Porto, Porto, Portugal

Pedro F. Mayuet
University of Cadiz, Cadiz, Spain

Eanna McCarthy
I-Form, Advanced Manufacturing Research Centre, and Advanced Processing Technology Research Centre, School of Mechanical and Manufacturing Engineering, Dublin City University, Dublin, Ireland

Greg McNamara
School of Mechanical and Manufacturing Engineering, Dublin City University, Dublin, Ireland

Tshepiso P. Molaba
Department of Chemistry, Nelson Mandela University, Port Elizabeth, South Africa and Polymers and Composites, Council for Scientific and Industrial Research, South Africa

Ilaria Papa
Department of Chemical, Materials and Production Engineering, University of Naples Federico II, Naples, Italy

Vijay Parmar
The Maharaja Sayajirao University of Baroda, Baroda, Gujarat, India

Federico París
Sevilla University, Seville, Spain

Piyush Patel
The Maharaja Sayajirao University of Baroda, Baroda, Gujarat, India

Sooraj Patel
Advanced Materials Processing Research Group, Indian Institute of Technology Gandhinagar, Gandhinagar, Gujarat, India

Alessia Serena Perna
Department of Chemical, Materials and Industrial Production Engineering, University of Naples Federico II, Naples, Italy

Bernhard L.A. Pichler
TU Wien, Vienna, Austria

Francisco M.A. Pires
Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal and Faculty of Engineering of the University of Porto, Porto, Portugal

Luís D.C. Ramalho
Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal

Bushra Rashid
Universiti Putra Malaysia, Serdang, Selangor, Malaysia and Middle Technical University, Alzafarany, Baghdad, Iraq

Daniel E.S. Rodrigues
Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal

Pietro Russo
Institute for Polymers, Composites and Biomaterials, National Research Council, Pozzuoli, Italy

Alexander A. Safonov
Skolkovo Institute of Science and Technology, Moscow, Russia

Jorge Salguero
University of Cadiz, Cadiz, Spain

Luca Sorrentino
University of Cassino and Southern Lazio, Cassino, Italy

S.P.B. Sousa
Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal

Catarina S.P. Borges
University of Porto, Porto, Portugal

Isidro J. Sánchez-Arce
Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal

Fabienne Touchard
Institut Pprime, CNRS-ISAE-ENSMA-Université de Poitiers UPR, Département Physique et Mécanique des Matériaux, ENSMA 1 av Clément Ader, Futuroscope, Chasseneuil, France

Fausto Tucci
University of Salerno, Fisciano, Italy

Alex Valadez-González
Scientific Research Center of Yucatan, Mérida, Yucatán, México

Robertt Valente
University of Aveiro, Aveiro, Portugal

Ana P. Valerga
University of Cadiz, Cadiz, Spain

Juan M. Vazquez-Martinez
University of Cadiz, Cadiz, Spain

Alexander Vedernikov
Skolkovo Institute of Science and Technology, Moscow, Russia

Yong Yuan
Tongji University, Shanghai, China

Jiao-Long Zhang
Tongji University, Shanghai, China and TU Wien, Vienna, Austria

EDITORIAL BOARD

Editor in Chief

Dermot Brabazon

I-Form, Advanced Manufacturing Research Centre, and Advanced Processing Technology Research Centre, School of Mechanical and Manufacturing Engineering, Dublin City University, Dublin, Ireland

Section Editors

Manoj Gupta, Section 1: *Metal Matrix Composite Materials*

Department of Mechanical Engineering, NUS, Singapore

Dermot Brabazon, Section 2: *Polymer Matrix Composite Materials*

I-Form, Advanced Manufacturing Research Centre, and Advanced Processing Technology Research Centre, School of Mechanical and Manufacturing Engineering, Dublin City University, Dublin, Ireland

Fatima Zivic, Section 3: *Ceramics Matrix Composites*

Faculty of Engineering, University of Kragujevac, Kragujevac, Serbia

Eva Pellicer, Section 4: *Smart Composite Materials*

Departament de Física, Universitat Autònoma de Barcelona, Campus de la UAB, Barcelona, Spain

Dermot Brabazon, Section 5: *Processing of Composite Materials and Physical Characteristics*

I-Form, Advanced Manufacturing Research Centre, and Advanced Processing Technology Research Centre, School of Mechanical and Manufacturing Engineering, Dublin City University, Dublin, Ireland

Robertt Valente, Section 6: *Design Methods for Composite Materials*

Center for Mechanical Technology and Automation, Department of Mechanical Engineering, University of Aveiro, Portugal

Mohamed El Mansori, Section 7: *Nature Based and Inspired Composite Materials*

Arts et Metiers Institute of Technology, Mechanics Surfaces and Materials Processing, HESAM Université, Châlons-en-Champagne, France
Texas A&M Engineering Experiment Station, Institute for Manufacturing Systems, College Station, TX, United States

Lorna Fitzsimons, Section 8: *Life Cycle Analysis and Sustainability of Composite Materials*

Advanced Processing Technology Research Centre, School of Mechanical and Manufacturing Engineering and the Water Institute, Dublin City University, Dublin, Ireland

Antonello Astarita, Section 9: *Joining of Composite Materials*

Department of Chemical, Materials and Industrial Production Engineering, University of Naples Federico II, Naples, Italy

PREFACE

This is the first *Encyclopedia of Materials: Composites* published by Elsevier which presents a vast and widely encompassing content in the area of composite materials science and engineering. Composite materials have become even more important and ubiquitous over the recent decades due to the many advantages that they can provide over single monolithic materials. This includes improvements in the properties such as the physical, electrical, chemical, optical and magnetic properties which can be achieved by combining two or more materials.

The two main types of composites, Metal and Polymer matrix based, are presented in detail within Sections 1 and 2 respectively while Ceramic matrix composites are presented in Section 3. Smart composites which is an area that is growing fast with increasing industrial relevance is covered in Section 4. Assessing the properties of composite materials thereby enabling their application is a crucial aspect of composite materials development and usage. As such, Section 5 presents the testing methods used and property results from the testing of composite materials. The design of composite materials is covered in Section 6. The recyclability and sustainability of materials used in products is an ever more important topic. There are some challenges to achieve well the recyclability of composite constructs. The Encyclopedia presented two Sections covering this one (Section 7) covering nature based composites and another covering the life cycle analysis of composite materials (Section 8). In the last section of the Encyclopedia, Section 9 covers how to join composite materials together and with more conventional monolithic materials.

As an Encyclopedia, these sections were prepared to be the primary central source of background knowledge for undergraduate, postgraduate and researchers studying or working with composites. The audience of this work covers both academic and industrial researchers. In today's composite materials market, engineers, architects, and even policy makers, need reference literature where to find definitions, concepts and state-of-the-art knowledge. As such this Encyclopedia will be an invaluable reference for engineers, architects, scientists, and policy makers.

Each section contains articles written by world experts in their area. As well as providing the latest background information, the state of the art in the niche areas is presented in the individual articles. A particular concern in preparing these articles by the authors and Section editors was to make the content as accessible as possible to the reader. This is important given the multi-disciplinary nature of people working on the development and implementation of composite materials.

I take this opportunity to thank the 337 authors from across the world who have contributed the 171 articles to this Encyclopedia. It has been enjoyable to work with you are encouraging to see your expertise, interest and desire to help others from your contribution. With the many co-authored articles, there has been extensive collaboration which has resulted in a more informed and well-presented Encyclopedia content for the reader.

I am indebted also to the members of the Editorial team who have worked many long hours over the last couple of years to provide feedback and iterate on articles with the authors. The Editorial team have collectively many years of expertise working in their research areas. This team was formed via a variety networking events including conferences such as ESAFORM and Global Conference on Nanomaterial Forming (Manoj Gupta, Robertt Valente, Antonella Astarita), EU research projects and COST Actions (Fatima Zivic and Eva Maria Pellicer), and via other Dublin City University and sustainable engineering networking events (Mohamed El Mansori and Lorna Fitzsimons). I thank the Elsevier Major Reference Works team who supported in a professional manner the compiling of this work. In particular, I thank Laura Jackson, Sajana P K, and Ruth Rhodes for their direction and support throughout the preparation of this Encyclopedia.

Dermot Brabazon
May 2021

Design Methods for Composite Materials: Preface

Robertt Valente, University of Aveiro, Aveiro, Portugal

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Composite materials were always a rich and challenging field, well-known to constantly pushing the boundaries of the state of the art in engineering and material sciences fields, in general, and design, modeling and numerical simulation areas, in particular. This interest from the scientific and industrial communities can be easily seen from the number of publications, patents and products that, directly or indirectly, deals or are affected by the knowledge in composite materials' area. Putting an extra focus on this consolidated importance, the articles chosen to be part of the Design Methods for Composite Materials section aim to present different views, methodologies, techniques and results on the current capabilities of modeling and simulation approaches, their advantages and limitations and, consequently, pave the way to further developments and achievements in this field.

Starting with the article co-authored by Carlone, Aleksendrić and Sorrentino, entitled "Neural based optimization of composite curing process", the authors cover in a comprehensive and integrated way the capabilities of applying nature based and inspired optimization approaches to the design, critical thinking and prototyping on thick and thin resin cured composite laminates. Their approach focuses on the challenges that emerge when combining artificial neural network with (genetic) optimization algorithms, particularly for the *a priori* determination of the best choice of process parameters for curing stages. The article provides also a good insight on the current state of the art in the field, rich discussions on the different strategies that can be adopted and conclusions that can be scaled to industrial applications.

With the article "3D topology optimization of continuous fiber-reinforced structures", by Safonov, the capabilities of additive manufacturing processes on obtaining 3D continuous fiber structures are explored. Particularly dedicated to topology optimization approaches when applied to the design stages of such structural components, the proposed algorithmic approaches are incorporated into Abaqus Finite Element (FE) software by means of a user material subroutine. From this well-known and widely spread simulation platform, the presented research and procedures are dedicated to finding the optimal distribution of material density as well as fiber orientations, focusing on the production of complex industrial parts. The article guide the reader in the theoretical and algorithmic aspects of implementation, therefore representing an added value for the numerical simulation community.

From a conventional FE approach to a more unconventional and innovative simulation methodology, on the article "Using a meshless method to predict the strength of adhesive single lap joints", Ramalho and co-authors explore the capabilities of the Radial Point Interpolation Method (RPIM), a meshless approach for dealing with unstructured and complex geometry cases, now applied to joining operations by adhesive bonding, particularly single lap joints. The article provides an extensive and comprehensive review on the state of the art in meshless methods over the time, guiding the reader into the evolution of this field. Additionally, theoretical and implementation aspects of RPIM are provided, together with detailed results for robustness assessment of the proposed approaches.

Still aligned with nonconventional modeling approaches, Rodrigues and co-authors present an alternative approach for multiscale homogenization aspects related to composite materials, in the article "Homogenizing the elastic properties of composite material using the NNRPIM". In their work, the capabilities and challenges of adopting the Natural Neighbor Radial Point Interpolation Method (NNRPIM), as a basis for homogenization procedures, is described in detail, as a way of applying a multiscale strategy for the determination of elastic properties of general heterogeneous materials. The article provides the basis for the construction of NNRPIM, together with details for implementation and, finally, a set of assessment benchmarks in evaluating the computational performance of the proposed approach, the robustness of the obtained solutions and the evolution potential of that numerical strategy, with a critical comparison between more conventional strategies.

Moving forward to production aspects, the article "Design criteria for pultruded structural elements", by Tucci and Vedernikov, provides insight and a detailed analysis of the potential of fiber reinforced polymers, together with the challenges associated to the production of real structural components by means of pultrusion manufacturing. Different design criteria are described, analysed and compared, in detail, making the article a reference work for beginners and experts in the field. A number of design variables are considered, and their interchangeable influence is also analysed, providing guidelines for the most relevant ones, as well as indications for further research and developments in the area.

Finally, and moving to a distinct set of composite materials, in the article entitled "Assessment of the added value of multiscale modeling of concrete for structural analysis of segmental tunnel rings", by Zhang and co-authors, the focus is put on concrete materials for demanding structural applications, and the challenge of providing modeling and numerical solutions by means of multiscale approaches. A detailed and careful introduction is provided on the particular aspects and challenges of such a class of materials, together with the needs from the point of view of the constitutive modeling and design for maximum mechanical and structural performance. Based on a real design and manufacturing case, the article guides the author in the process of decision making, theoretically and practically grounded, and particularly focusing on the proper and cost-effective determination of the material properties using multiscale strategies.

All together, these articles cover different modeling and simulation strategies, distinct materials and applications, each one with its particular challenges and potential for future developments in the robust and integrated design field.

Neural Based Optimization of Composite Curing Process

Pierpaolo Carlone, University of Salerno, Fisciano, Italy

Dragan Aleksendrić, Faculty of Mechanical Engineering of the University of Belgrade, Belgrade, Serbia

Luca Sorrentino, University of Cassino and Southern Lazio, Cassino, Italy

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Introduction

Nature has always been a source of inspiration for human beings. Apart from the obvious influence on human behavior, this evidence can also be found in noble areas, such as art and science. Learning from nature and then elaborating or engineering is what scientists have been doing for centuries and, it is apparent that most of the concepts recalled in this article can be easily associated with this main scheme.

The idea itself of “composite material” is an excellent example of this sort of inspiration. In fact, wood is a natural material based on the combination of two polymer species, namely cellulose fibers reinforced inside the resinous matrix of polysaccharide lignin (Harris, 1999). Since ancient times, mixing or combining different materials has been a common strategy to achieve an enhancement in performance, however, a rigorous approach to science and engineering of composite materials can be roughly dated back to the 1950s (Harris, 1999).

In material science, the classification of a material as a composite is extremely general and, in some cases, excessively exploited. Composites are primarily classified by considering the matrix material. From this point of view, they are distinguished in metal-matrix composites (MMC), ceramic-matrix composites (CMC), and organic-matrix composites (OMC). The latter category includes both polymer-matrix composites (PMC) and carbon-matrix composites (generally introduced as carbon-carbon composites CCC). A further classification is based on the type of reinforcement: distinguishing particulates, whiskers, (discontinuous and continuous) fibers, and woven textiles (Isaac and Ori, 1994). Being the methodology described in the present article specifically applied to thermoset based PMC, hereafter the term composite materials or composites or PMC is intentionally restricted to that particular class.

Composites exhibit distinctive advantages compared to monolithic materials. Excellent specific strength and stiffness, significant fatigue resistance, and the ability of a material to be adapted or tailored at the design stage for the specific function are clear examples. Further improvements are anticipated in corrosion behavior (Miracle and Donaldson, 2001; Campbell, 2010) as well. The development of dedicated theory to predict the structural behavior, as well as the advances in computer and software, allowed scientists and engineers to tailor and control the properties of composites by controlling their ingredients and distribution. Indeed, the proper selection of material, volume fraction, shape, and architecture of what is referred to as secondary or reinforcing phase, often simply indicated as reinforcement, as well as its combination with the primary or continuous phase, namely the matrix, provide engineers with novel materials and design flexibility (Campbell, 2010). Due to these reasons, the incorporation of PMC has increased in various industries ranging from aerospace, automotive, naval, energy sector to sporting commodity.

In a broad sense, advanced composites are already optimized materials, since they are ad hoc conceived consisting of a well-defined properties balance required for the particular set of applications (Isaac and Ori, 1994). Significant literature has been devoted so far to apply sophisticated optimization algorithms to various material configuration in order to provide the requested answer in terms of static and dynamic behavior, natural frequencies and vibration modes, impact loading, etc. Nevertheless, the achievement of enhanced and tailored properties cannot disregard precise manufacturing concerns. Certainly, the necessity for opportune integration of various materials and a well-defined shape formation poses strong constraints on the selection of suitable manufacturing processes and the appropriate tuning of process parameters.

In concept, PMC manufacturing is based on three fundamental steps: impregnation, consolidation or shaping or forming, and solidification or curing. The indicated steps must not be assumed as strictly consequential, indeed, in some process, two or eventually all of them can be achieved at the same time. While forming operations are intuitively targeted to the definition of the geometry, dimension, and tolerances of the final product, the impregnation step is strictly related to the combination of the primary and secondary phases. This combination can be promoted off-line, as in the case of prepregs, or on-line, as typically happens in “wet” processes like wet filament winding and pultrusion. In both cases, the final aim is to materialize an intimate contact between matrix and reinforcement, in order to guarantee adequate mechanical properties and failure mechanisms.

Excluding manual processing, such as hand or spray layup, where optimization procedures are hardly justifiable, a variety of manufacturing techniques has been proposed and effectively applied for composite production. The most used methods are vacuum bagging and autoclave curing, liquid composite molding processes, continuous processes (filament winding and pultrusion), and compression molding. In recent years, automated tape and fiber placements methods have been developed as well. Obviously, a detailed description of PMC manufacturing processes is beyond the scope of this article. In what follows the fundamentals of adopted methods for PMC manufacturing are recalled. Readers are invited to explore the details in Gutowski (1997) and the therein cited literature.

Autoclaves are pressure vessels equipped with heating and cooling facilities and providing the possibility of a localized vacuum application. Autoclave processing involves the lamination and vacuum bagging of the processing and ancillary materials on the surface of a forming die, which is then transferred to the autoclave. Work pieces are positioned inside the autoclave with the

vacuum facility. The curing process is designed considering the pressure and temperature cycles. The combination of these factors provides enhanced mechanical properties depending on three aspects: material consolidation, complete polymerization of resin, and voids reduction or suppression. After the curing cycle, composite material is demolded, inspected and assembled. The term liquid composite molding processes include a class of manufacturing methods based on the dry fibrous preform impregnation injecting or infusing an opportunistically catalyzed resin. The preform is shaped and closed between solid-solid or solid-flexible dies, depending on the nature of the applied driving pressure. The impregnated preform is then heated to activate or accelerate the cure reaction. Prime advantages of the LCM processes include the ability to manufacture complex shapes products with excellent precision and flexibility of the reinforcement architecture and volume fraction, better surface finishing, and the less direct operators' intervention on the processing reactive resin. In pultrusion, reinforcing fibers pulling is carried out through some guiders. This is followed by the impregnation of fibers with the resin in an open bath or by using an injection chamber. The impregnated reinforcement is further forced through the heated die. Inside the die, the exothermic cure reaction of the thermoset resin is activated upon receiving the heat provided through the electrical heaters or hot oil. Consequently, the material changes its states from reactive liquid to gel and eventually to vitrified solid. The process is concluded with the pulling out action of all the cured and solidified products by employing reciprocating or caterpillar mechanical systems followed by the cutting of product in required lengths. In the case of the filament winding process, initially continuous reinforcement rovings or woven textile layers are carried out which are then impregnated by catalyzed resin and are finally wound around a rotating mandrel following a geodesic path. Undesired displacement of the fibers and modification of reinforcement architecture is generally prevented through pre-tensioning the fiber while wiping systems remove the excess resin. After the deposition, the composite undergoes cure, at room or high temperature, depending on the resin system. Comprehensive process descriptions are available in [Gutowski \(1997\)](#).

Compression or press molding involves the forming of the composite material by means of the closing force exerted by two die halves which are mounted on a press. As a result, the induced compression impels the cavity filling action by raw material. It offers better precision in dimension and surface finish at high production rates. Work piece solidification is then achieved activating the cure reaction by heating the die.

Irrespective of the adopted manufacturing technique, the curing process is a common and crucial step in PMC production. In a simple explanation, the curing cycles possess resemblance to the heat treatments containing single or more heating and holding stages along with an eventual (slow) cooling to room temperature ([Gutowski, 1997](#)). Additional complexities are related to phase changes occurring into the reactive resin. The way this thermal cycle is applied to the material is dependent on the process. In the case of autoclave curing, the rate with which gradual enhancement in the autoclave temperature is realized depends upon the physical properties and thickness of the processing part. The enhancement in temperature is achieved by utilizing the hot recirculating air or other gases. Similar strategies are adopted for oven curing of filament wound parts or work piece realized by LCM. Differently, in pultrusion, the local temperature is controlled to well-defined levels, while the heating rate and the exposure time at each temperature depend on the pulling speed.

The design of curing stage presents not negligible difficulties raising from physical and chemical irreversible phenomena involved ([Isaac and Ori, 1994](#)). In industry, generally, the trend is to apply the cure profiles suggested by the suppliers. In this article, the curing cycle definition by optimization algorithm and neural-based techniques is discussed.

Curing Process

The curing process is necessary to transform the matrix from a viscous liquid to a solid, promoting the crosslinking among monomers. To allow matrix curing a heating cycle is needed, as the resin kinetic is controlled by the temperature, but this thermal cycle can induce different kinds of damage and defects in the part being manufactured; therefore, to design a suitable cure process is essential for avoiding such issues, that influence the reliability and the strength of the parts. Therefore, an accurate time-temperature profile should be precisely designed in order to achieve the right conditions of the degree of cure ([Shevtsov et al., 2012](#)).

Among the cure-induced defects, uncured resin and resin degradation are typical of high thickness laminates, that present a thickness higher than 10 mm. In fact, the uncured resin is an issue presented because of the associated lower thermal conductivity of the resin. Generally, for the resin reaction activation, the heat supply is needed. However, the temperature increase in the inner part of resin(core) is challenging considering the lower thermal conductivity of the resin that hinders the heat from reaching the laminate core correctly. This results in causing a decrease in the structural characteristics of the part. In concern with the issue of resin degradation, the low thermal conductivity again poses difficulty as the resin degradation induced by the exothermic peak. In fact, the heat developed by the resin polymerization, that is an exothermic reaction, is not able to reach the surface of the laminate, and so it accumulates in the laminate center, making the temperature rise and causing the resin deterioration ([Kondyurin et al., 2012](#)). In general, the resin and prepreg manufacturers provide thermal cycles that are suitable for standard thickness laminates, that is 5 mm, but these recommendations are not suitable for high thickness laminates. In a work of [Esposito et al. \(2016\)](#), it is showed that a manufacturer's thermal cycle can induce a very high thermal peak in the material, in fact in a 25 mm thick laminate a temperature 75°C higher than the cycle temperature was found. The strength decrease induced by thermal peak has been the topic of several studies ([Esposito et al., 2016](#); [Olivier and Cavarero, 2000](#); [Nightingale and Day, 2002](#); [Sorrentino et al., 2017](#)), and a decrement of 17% in the interlaminar shear strength was found by [Sorrentino et al. \(2017\)](#). The increase of the temperature in the center of the laminate causes an uneven cure degree trend along the thickness of the laminate, that, in turn, lets residual stress arise. In fact, the higher temperature boosts the cure rate and so the resin in the center polymerizes before than that of the outer

zones. This is a problem since the resin shrinks during polymerization, and so the differences in the resin shrinkage let internal stress appear.

Other problems are due to the deformations induced by the cure that are not harmful to the strength of the manufactured parts but cause the part rejection due to assembly issues. Indeed, manufactured parts have tight dimensional and geometrical tolerances, necessary to allow the assemblage in final products, preventing aesthetical and, in some cases, structural defects. An issue quite common in the aeronautical field is the shimming, that is a time-consuming operation and causes the increase of aircraft weight and production costs (Kappel *et al.*, 2013). In fact, the shimming consists in filling the gap between two mating surfaces, and it is articulated in different steps: at first, the space between the parts to be assembled must be gauged; then, thin slabs are machined considering the space to be filled; finally, the produced shims are bonded in the part. During the curing process, there is a generation of the residual stress which causes the part distortions and are induced by several events that arises in the curing process. It is worth remembering that the residual stresses consist of self-balanced stresses that are due to intrinsic factors and are not induced by external loads (Ding *et al.*, 2016a). Several factors induce residual stresses among them, the anisotropy of composite material and the interaction with the mold are prominent (Ding *et al.*, 2015). There are two main kinds of residual stress deformations: the warpage and the spring-in. In detail, the warpage is a flat laminate warping, while the spring-in is the alteration of the angle between two flanges of an L-shaped laminate. The former is typical of flat laminates, while the latter of curved ones; however, in general, a part presents both curved and flat zones, so both kinds of distortion coexist in the same part.

Among the several factors that induce residual stress in a composite material laminate, material's CTE (Coefficient of Thermal Expansion) value is one of the most important. The CTE of the matrix is different from that of the fibers, that for the carbon fibers is even negative, and this aspect give rise to different types of residual stresses. Ones arising at a local scale level, between fiber and matrix, may provoke delamination and cracking, so the laminate failure, but they are not able to cause distortion since their effect is dissipated through the laminate thickness (Ding *et al.*, 2016b; Sorrentino and Bellini, 2015a). Considering the single ply of a laminate, its CTE is transversely isotropic, consequently distortion may rise at this level. Indeed, to obtain a deformation of the whole laminate the structural neutral axis of the laminate should not coincide with the geometrical one, otherwise these stresses do not affect the shape. This condition is verified for curved laminate and for laminate with an unbalanced or unsymmetrical stacking sequence, while the residual stresses are self-equilibrated in the flat laminates having symmetry and balance in the stacking sequence. However, for all kinds of laminate (flat or curved, balanced or unbalanced, etc) the in-plane CTE is lower than the through thickness one, and in such case residual stress give rise to deformation (Mahadik and Potter, 2013).

An effect similar to that one of the CTE is exerted by the resin cure shrinkage: the fiber does not cure and, consequently, their chemical shrinkage is null; therefore, laminate displays lower in-plane chemical shrinkage in comparison with the through-thickness one, since in this latter direction the effect of the fiber is negligible (Baran *et al.*, 2017). The effect of both CTE mismatch and chemical shrinkage depends on the resin content, so the gradient of fiber content caused by the resin bleed during the compaction phase, at the beginning of cure cycle, may amplify the impact of the previous factors, especially for flat laminates, in which it accentuates the warpage (Kappel *et al.*, 2011).

Another cause for the cure induced residual stresses and distortions is the interaction between the laminate being cured and the mold (Kappel, 2016). In fact, the CTE of the mold material is different from that of the laminate, and the different thermal deformation during the cure cycle are sufficient to induce tension stress in the laminate (Bellini *et al.*, 2014). A brief account of some phenomena occurring in the cure cycle is necessary to understand this kind of residual stress: during the first part of the cycle the temperature rises, so the mold expands; at that time, the resin cannot sustain shear stress since the degree of cure is still too low. Therefore, the most stressed composite ply is the one in contact with the mold surface, and the others experience less stress. As the polymerization proceeds, the resin shear modulus rises and the uneven stress field, which was generated during heat ramp, remain intact in the laminate. Upon the laminate extraction from the mold, the stresses release and the distortions become evident. This phenomenon is intensified in the vacuum bag process because the bag is flexible and so the layer in contact with it is not highly stressed compared to the layer touching the mold. Moreover, the vacuum bag give rise to an uneven distribution of the fiber content, so the effect of the curing process on the distortion is amplified, as described above.

Cost-saving achieved by replacing the comprehensive experimental activities with numerical simulation has been recognized so far. This applies both to PMC product as well as process design that is demonstrated by the scientific literature (Sorrentino and Bellini, 2015b; Carlone *et al.*, 2018). Numerical models are suitable to calculate the temperature, the degree of cure and the induced deformation, but they need some information, as the material thermal properties, the kinetic properties of the resin and the thermomechanical behavior of the materials involved (laminate and mold), besides the boundary conditions, that are the heat cycle and the blocking conditions of the laminate inside the mold. The determination of the material properties is a fundamental fact for the numerical simulation of the process since the results can be heavily influenced by the material properties. To generate a complete cure process simulation, two models are necessary: a thermochemical model to determine the temperature and the degree of cure, and a thermomechanical model to determine residual stress and consequent deformation. However, if the target is to verify if temperature peak arises during cure, the former model is sufficient, without the need of implementing the latter. The thermochemical model is based on the heat transfer equation (Kappel *et al.*, 2013; Sorrentino and Bellini, 2016):

$$\rho_c c_{p,c} \frac{dT}{dt} = \nabla(k_c \nabla T) + \rho_r v_r \dot{Q} \quad (1)$$

In that equation, T is the temperature, t the time, ρ the density, v the volumetric content, c_p the specific heat and k the thermal conductivity. The subscript c and r refer to the composite and the resin, respectively. The density of composite material is

determined as the weighted average on the volumetric content of fiber and matrix, while the specific heat as an average weighted on the mass content of fiber and matrix (Sorrentino *et al.*, 2009, 2015, 2014). The composite material thermal conductivity is considered as anisotropic property, due to its dependence on the fibers direction, and there are various models to define the conductivity along the different directions. For example, the conductivity along the fiber for a unidirectional ply can be calculated from that of fiber and matrix through an average on volumetric content, while that perpendicular through the Nielsen equation:

$$\frac{k_c}{k_r} = \frac{1 + ABV_f}{1 - B\psi V_f}, \quad A = k_E - 1, \quad B = \frac{(k_f/k_r) - 1}{(k_f/k_r) + A}, \quad \psi = 1 + \left(\frac{1 - V_{\max}}{V_{\max}^2} \right) V_f \quad (2)$$

There is a possibility to carry out the calculation of heat rate $\dot{Q}$ produced by the polymerization reaction as the product of the total reaction heat H_r with the cure rate $d\alpha/dt$. There are different models to calculate this rate, but the most frequently employed is the autocatalytic one:

$$\frac{d\alpha}{dt} = A e^{\frac{-E}{RT}} \alpha^m (1 - \alpha)^n \quad (3)$$

In which A is the frequency factor, E the activation energy, R the universal gas constant and m and n represent the reaction orders. To determine the coefficient of Eq. (3) some DSC tests are needed, according to the ASTM E2070 standard.

In case of proceeding with the thermomechanical step simulation, it must be remembered that at the beginning of the polymerization resin is liquid, then it reaches the gelation point and become rubber and finally a solid, in the vitrification point. The first point is identified with a precise cure level, usually the 25%–30%, while the latter is reached when the laminate temperature exceeds the glass transition temperature T_g , that can be calculated using Di Benedetto equation:

$$T_g = \frac{\lambda \alpha (T_{g\infty} - T_{g0})}{1 - (1 - \lambda) \alpha} + T_{g0} \quad (4)$$

Where λ is a characteristic coefficient, T_{g0} is the minimum glass transition temperature and $T_{g\infty}$ the maximum one. Different mechanical models have been proposed to simulate the residual stresses, but the most frequently adopted are elastic and viscoelastic. Chemical shrinkage arising in the resin is supposed to vary in linear fashion with the degree of cure, whereas in the case of the thermal expansion the linear dependence from the temperature might be considered. The classical laminate theory can be adapted to compute the residual stress as a function of all the phenomena happening during the curing cycle.

Simulation tools are mainly based on hard computing techniques, namely finite element method (FEM), finite difference method (FDM), and finite volume method (FVM). Generally speaking, these methods are able to provide a reasonably accurate approximation of the ideal solution to the formulated problem, depending on discretization and truncation errors (Tucci *et al.*, 2020). In the interest of conciseness, the following section recalls only the main idea, readers are invited to explore the dedicated literature for deeper understanding. As a common point, the generic problem under consideration is a mathematically formulated set of partial differential equations that describe the governing equations (such as balance or equilibrium) of the modeled phenomena existing in defined space and time domain and in accordance with the imposed initial and boundary conditions. Then, depending on the method, the solution is estimated either by:

- (1) Replacing derivatives with finite quotients and then adopting matrix algebra methods (in FDM);
- (2) Assimilating a continuous domain to geometrically simpler and non-overlapping discrete entities and assuming the weak formulation of the problem as the starting point (in FEM);
- (3) Decomposing the domain into finite or control volumes (so-called dual mesh) and solving, for each volume, the governing equations in terms of conservation laws (FVM).

Other approaches involve boundary element methods and meshless analysis (Bogetti and Gillespie, 1992).

The cited methods proved their capability in accurately reproducing the process in a virtual framework. Nevertheless, simulations can be computationally costly and time-consuming. In the next section, an alternative approach, based on the coupling of hard computing, soft computing, and optimization algorithms is discussed.

Neural Based Curing Process Optimization

Process Optimization

A considerable amount of research attempts is focused on the cure cycle optimization (Aleksendrić and Carlone, 2015; Carlone *et al.*, 2014; Vasudevan, 2009). Specifically, when composite curing is considered, the formulation of the prime objective function is based on the time and cost involved in processing. In addition to that, it can consist of the arising residual stress or the shape change/degradation (Aleksendrić *et al.*, 2010; Hossain *et al.*, 2016). Technological constraints, related to exceeding temperature, partial curing, and variation in temperature or cure are taken into account introducing opportune penalties (Struzziero and Skordos, 2017). The initial research focused on the early attempts were based on inefficient and expensive trial-and-error methods, which did not ensure the attainment of an optimized set of parameters (Aleksendrić and Duboka, 2007, 2006; Aleksendrić and Barton, 2009). Moreover, it is not generally applicable and more of the case-specific with inhibited applicability (Aleksendrić *et al.*, 2010; Aleksendrić, 2010; Aleksendrić and Senatore, 2012). Rigorously speaking, such attempts should be more correctly referred to

as a refinement of sub-optimal solutions rather than optimization in the strictest meaning of the term. In the case of considering the mathematical or computer science, the optimization problem is defined to search the minimum (or maximum) point of a function $f: R_n \rightarrow R$, where, in R_n admissible set of solutions are obtained. The past decade observed the introduction of numerous algorithms which were further implemented and improved. Along with that, the relevant applications to the composite design and manufacturing have been described in the literature.

Due to space limitation, a comprehensive analysis of such optimization procedures is not considered as the main focus of this work. Additionally, in the authors' point of view providing the detailed descriptive account of the mathematical base and robustness of optimization methods is beyond the scope of this work. In the sake of understanding and completeness, however, highlighting the difference between the derivative and derivative-free methods. Precisely, the derivative methods are also called the first order or gradient-based methods that are established on developing the considered function gradient. On the other hand, the derivative-free methods also known as zero-order method involves either the evolution of function value or carrying out the derivative approximation (Kolda *et al.*, 2003; Hvattum and Glover, 2009). In the case of optimization of engineering issues coming from the composite manufacturing field, different class of methods known as the direct search methods are frequently utilized. The most used algorithms are described as follows:

Simplex method

It is essential to first define the term simplex before proceeding with the method description. Simplex is the convex geometrical establishment in R_n . It is characterized by n number of dimensions, $(n + 1)$ vertexes and edges, whose dimension is $(n-1)$, in R_n . The quintessential examples include the triangle in the two-dimensional space R_2 and the tetrahedron in three-dimensional space R_3 . The simplex optimization was initially conceived by Spendley *et al.* (1962) in which further changes were implemented by Nelder and Mead (1965). According to this method, each vertex of the simplex is a candidate solution. The simplex changes its configuration at each iteration (walk) replacing one or more vertices (representing relatively worse solutions) by reflection, contraction, as well as expansion operations. Some strategies for avoiding the local minima, especially in multi-dimensional problems, have also been suggested.

Simulated annealing

Another algorithm based on providing the optimization that imitates the behavior of metals upon slow cooling. The basic idea consists of the progressive reduction of the temperature and molecular movements, i.e., the objective function unless the attainment of the lowest energy state i.e., the global minimum. In a likewise manner, the algorithm creates a solution at each iteration, and its acceptance depends on fulfilling the Metropolis criterion, up to the extent of satisfying the convergence criterion. The parameters that define this Metropolis criterion sharply influence the effectiveness of the Simulated Annealing algorithm in determining the searching space which can be of interest along with avoiding reaching the local minima. Extremely high temperature indicates acceptance of inopportune movements or repeated acceptance of reflected vertices and simplex stall. Whereas, low temperatures, or rapid cooling, inhibits the escaping capability of the simplex, penalizing algorithm convergence and quality of the solution. Few applications to composite manufacturing have been attempted in literature since the algorithm parameters tuning is with high complexity and precisely case specific.

Genetic algorithm

Genetic Algorithms (GA) falls into the computational model category that derives the inspiration from the natural selection and evolution. The algorithm work on first encoding the prospective solution of the considered problem on the data structure that resembling the chromosome structure. After that few recombination operators are employed for containing the integral information intact. In the concept of GAs, if we consider a certain population of individuals to be the representative of the potential solutions of the considered problem, they can be characterized by providing the fitness score which determines the quality or performance of the solution. From the fitness score, the individual with a higher score, i.e., highly fit individual will attain the reproduction possibility through the selection and cross over with an objective of creation of new individuals (offspring). On the other hand, the least fit individuals will obtain lesser opportunities in reproduction and eventually they die out (Bansal *et al.*, 2013; Yegnanarayana, 2006). Stochastic mutation of chromosomes is also possible. Eventually, after the creation of successive generations, there is a possibility of population evolution and attainment of the optimal solution.

Particle swarm optimization

First developed by Kennedy and Eberhart (1995), the Particle Swarm Optimization method is related to the simulation of social behavior. As intuitive, the method is inspired by the simultaneous and coordinate movement of birds or fishes composing a swarm. According to the algorithm, a set of particles (candidate solutions) moves to explore the search space. Each particle is firstly defined by its position (process parameters in this case). The movement of each particle is promoted defining a velocity that depends on the surrounding particles and the current best solution. In this way, the swarm tends to converge toward the intriguing domain area.

Fuzzy logic

Fuzzy logic systems do not rely on predictive mathematical model of process (Hossain *et al.*, 2016; Hu *et al.*, 2017; Dey *et al.*, 2017; Muthuramalingam *et al.*, 2014). Indeed, the fuzzy logic controller (FLC) exerts its actions in the form of linguistic rules,

i.e. membership functions, fuzzy rules, and rule interpretation, mimicking the human reasoning. These rules have to be combined into the fuzzy set theory simulating the behavior of the control expert. Defining of proper linguistic rules relies on the knowledge of the process to be controlled, but arbitrary choices (for example of the shape of membership functions, e.g., triangular, gaussian or trapezoidal) have to be made in the translation of control rules into the fuzzy set theory framework (Rubino *et al.*, 2016; Ross, 2010).

Recently, the procedures integrating numerical models and optimization algorithms have been the focus of a number of research efforts (Sorrentino and Bellini, 2016). Indeed, the involved aspects of complexity often inhibit the attainment of a closed-form solution that can potentially connect the process parameters with the end product or the process performance. Furthermore, in the case of employing the simplified assumptions, there is a challenge of decreasing the effectiveness and the relevance of the analytical model. On the other hand, simulation-based optimization strategies further interfaced with on-line monitoring systems (Rubino *et al.*, 2016; Aleksendrić *et al.*, 2016) along with active regulation devices, possesses the potential of applicability in the scheme of active process control (Struzziero and Skordos, 2017; Aleksendrić, 2010).

Considering the computational effort required by the iterative solution of the formulated problem, i.e., assuming different combinations of working parameters, other strategies, based on soft computing methods, are currently explored. Among others, artificial neural networks seem a promising technique to dramatically reduce simulation time.

Artificial Neural Networks

Artificial neural networks are soft computing tools having the ability for the approximation of nonlinear behavior of the dynamic process (Rai and Pitchumani, 1997). In detail, the behavior associated with these tools is inspired by the nervous system working. The interesting attribute of the neuron networks is the adaptiveness since the problem solving via programming precedes the "learning by example" (Aleksendrić and Carlone, 2015; Jahromi *et al.*, 2012). There is a scope of introducing the training for performing the considered function through regulating the weight factor values between the neurons. This can be achieved by either providing the information from outside or through the neurons themselves. The pattern recognition and learning are the integral part of the neural network learning and memory (Aleksendrić and Carlone, 2015; Carlone *et al.*, 2014; Jahromi *et al.*, 2012; Carlone *et al.*, 2013; Lawrynczuk, 2011). The dynamic type of neural network is stronger than the static neural network since it contains a memory part that aids in remembering the previous values and states of the network (Lawrynczuk, 2011). Specifically, the dynamic neural network depends on the present and the previous input values, along with the output values of the network. Hence, the gradient computation is carried out in a more complex way in comparison with the static network. The advantage is that the similar learning algorithms used for static network training can be employed to train the dynamic network.

Dynamic neural networks are distinguished from the other ways in regard to its general classification into Feed-forward Time-Delay Networks, and Feedback or Recurrent Networks. Specifically, in recurrent network the outputs of any particular neurons are diverted (fed-back) to neurons in the precedent layers.

Considering the present work, the application of neural-based optimization on the composite curing has been exploited based on the coupling of two different approaches, namely; ANN with general algorithms and fuzzy logic controllers.

Neuro-Genetic Based Optimization

The neuro-genetic based optimization procedure is structured on the coupling of hard and soft computing techniques with an aim of time and cost reduction. Specifically, a FE model was incorporated for the generation of a data set that in turn was utilized to train and test the neural model. Furthermore, this ANN was the part of a genetic routine (Aleksendrić and Carlone, 2015), as depicted in the schematic in Fig. 1. The cure of graphite-epoxy composite was simulated in order to test the proposed method. Further characterization was carried out by macro non-homogeneity, e.g., volumetric fraction local variations or boundary conditions. The work piece model illustrated the doubly curved complex shape that corresponds to a race car body foremost part (Aleksendrić and Carlone, 2015). The dimensional outline of a part was $750 \times 360 \times 260 \text{ mm}^3$, with a constant thickness equal to 30 mm. Unidirectional carbon fiber reinforcement was incorporated into the epoxy resin (Hercules AS4/3501-6) matrix. Physical properties of reinforcing phase and matrix as well as reaction kinetics and parameters of the resin are available in (Young, 1995). The work piece was initially placed on a rigid die for forming and was subsequently placed into a vacuum bag for the autoclave process. The vacuum bag and other tooling were accounted in the model depending on their equivalent heat transfer coefficients ($30 \text{ W/m}^2 \text{ K}$ for flexible bag and $80 \text{ W/m}^2 \text{ K}$ for mold at bottom surface) which were responsible for all the thermal resistances (Aleksendrić and Senatore, 2012). Considered heating rates and holding temperatures were varied from 0.25°C to $5^\circ\text{C}/\text{min}$ and from 70°C to 240°C , respectively. The prediction of either full or partial cure was provided upon process end which was based on specific cycle.

Firstly, the procedure was applied to estimate the autoclave thermal cycle resulting in a wanted degree of cure profile in selected locations, as in Eq. (5):

$$f(x, t) = |\gamma_W - \gamma_P| \quad (5)$$

In Eq. (5), x represents the autoclave temperature optimal value, t is time, and γ_W and γ_P represents the are wanted and predicted values of degree of curing (DoC).

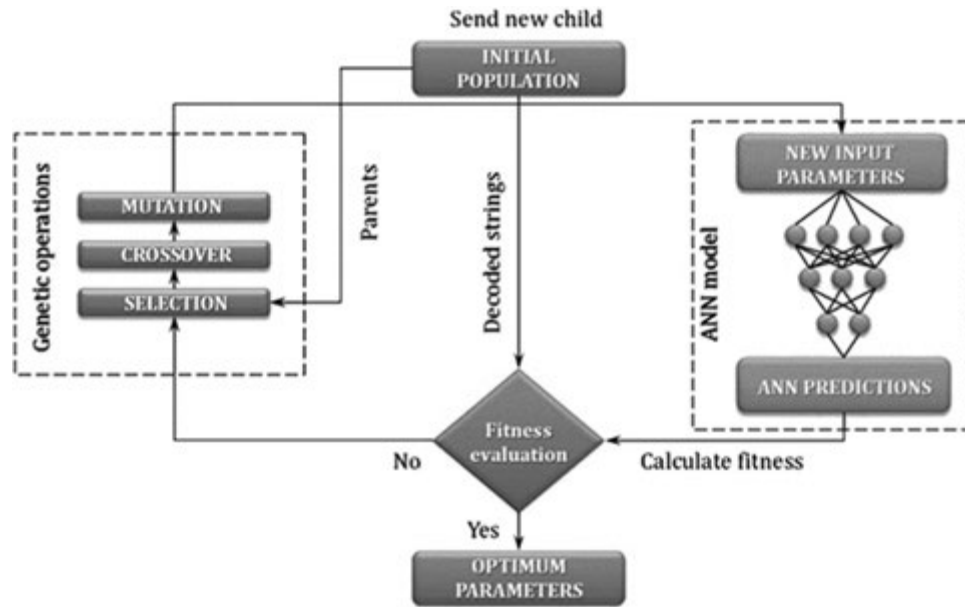


Fig. 1 The neuro-genetic based optimization model.

During the thermal cycle, genetic algorithms are applied to find out the autoclave temperature optimal value through achieving the reduction in the difference between desired and predicted DoC values. The prime objective of such an arrangement during the occurrence of successive generations of individuals is to minimize the DoC value difference up to zero. Upon attaining the fitness function value up to the determined criteria, the optimal autoclave temperature will be attained. Single optimization cycle was performed over 80 generations, using a “double vector” as population type with a size of 50 individuals. The “rank” calling function was used in combination with “tournament” selection function and reproduction function with 2 elite children. The crossover fraction was equal to 0.8. A two-step thermal cycle was considered as reference cycle.

The result of this calculation is reported in **Fig. 2**, comparing the desired and “suggested” profiles. The optimized autoclave temperature for the time duration of 30 and 120 min was under consideration. Optimized autoclave temperature T_{opt} was found to be less than the temperature T_h obtained by simulation with a maximum variation of 12°C (**Fig. 2**). It is reflected in the simulated DoC change. The optimization model returned the optimized autoclave temperature with an observed offset of 5°C.

Optimization model was also tested to find the optima autoclave temperature to obtain the 100% DoC between 100 min from the beginning of the cycle and the end. Considering the same time duration, the DoC calculated by FEM approached the 90% and correspondingly the autoclave temperature alteration was observed that is highlighted yellow as shown in **Fig. 3**. For instance, according to the neuro-genetic model, in order to enhance the DoC level from 90% to 100%, the composite material should be heated further for the subsequent 20 min as evident from the upward trend in the “ T_{opt} ” curve (light gray zone in **Fig. 3**). In order to achieve the maximum level of 100% DoC, the autoclave temperature should be increased by 60°C between 100 and 120 min. The curve “DoC_{simulated}” is the simulated output obtained by the dynamic neural model (change of DoC) developed using the optimized autoclave thermal cycle as the input. The simulated thermal cycle indicated the second heating start at 140 min (based on curve T_h), however, the interesting detail is that the earlier initiation of additional heating set up at 100 min (as illustrated by curve T_{opt}) resulted in the 60°C higher temperature i.e., 200°C instead of 140°C. In the imposed thermal cycle, this has a direct effect on the DoC as it attained the 100% at 150 min. Hence, the time duration between 150 and 240 min indicates the reminder of time which resulted in the time and cost-saving of the overall curing cycle. The further T_{opt} variation is not under the consideration due to 100% of DoC level obtainment. As a summary, it can be concluded that by imposing this thermal cycle, a large extent of time and cost-saving, and achievement of maximum DoC is possible in the curing procedure. This is achievable through employing the dual stage heating cycle with such optimized temperature variation.

Neural-Fuzzy Based Optimization

Intriguing capabilities of neural-based optimization are more evident when a real-time decision is required. This, for instance, is necessary in the case of an accident or unpredicted event causing a difference between the expected (already optimized cycle) and the detected evolution in material temperature and DoC. The ANN-based control strategy coupled with FLC was applied to tackle challenges such as huge growth in the controlled system complexity due to continuous technological advancements, and the rigorous requirement of achieving improved quality indicators in the controlled process (Tirian *et al.*, 2014). Neural networks and fuzzy systems extend the opportunity of solving a problem when a mathematical model doesn’t exist for a given problem. The

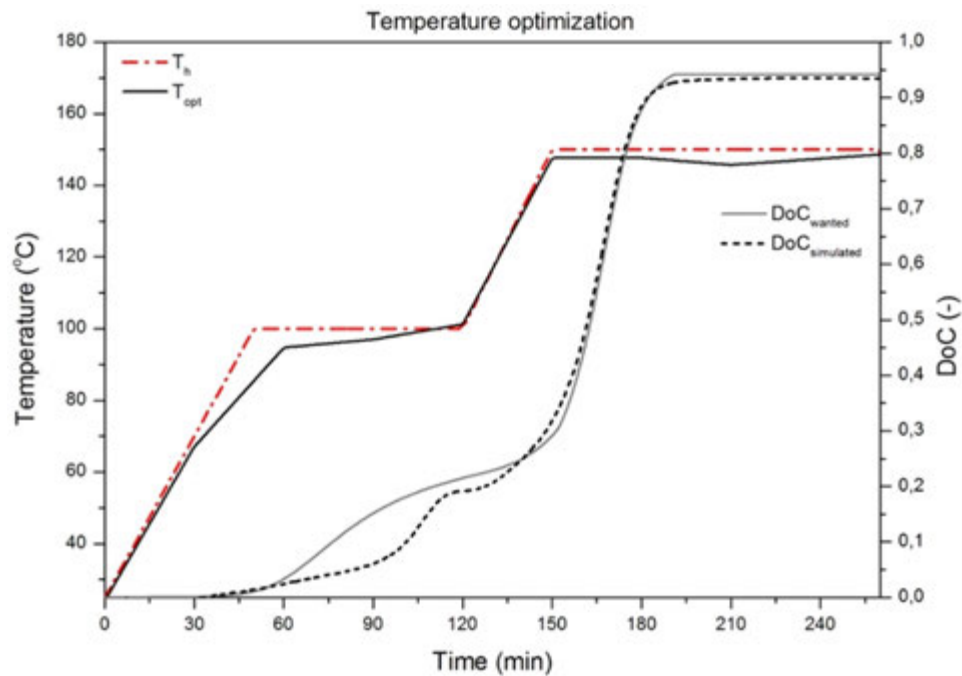


Fig. 2 Optimization of the temperature cycle of autoclave process (case of two-steps thermal cycle). Reproduced from Aleksendrić, D., Carlone, P., Čirović, V., 2016. Optimization of the temperature-time curve for the curing process of thermoset matrix composites. *Applied Composite Materials* 23, 1047–1063.

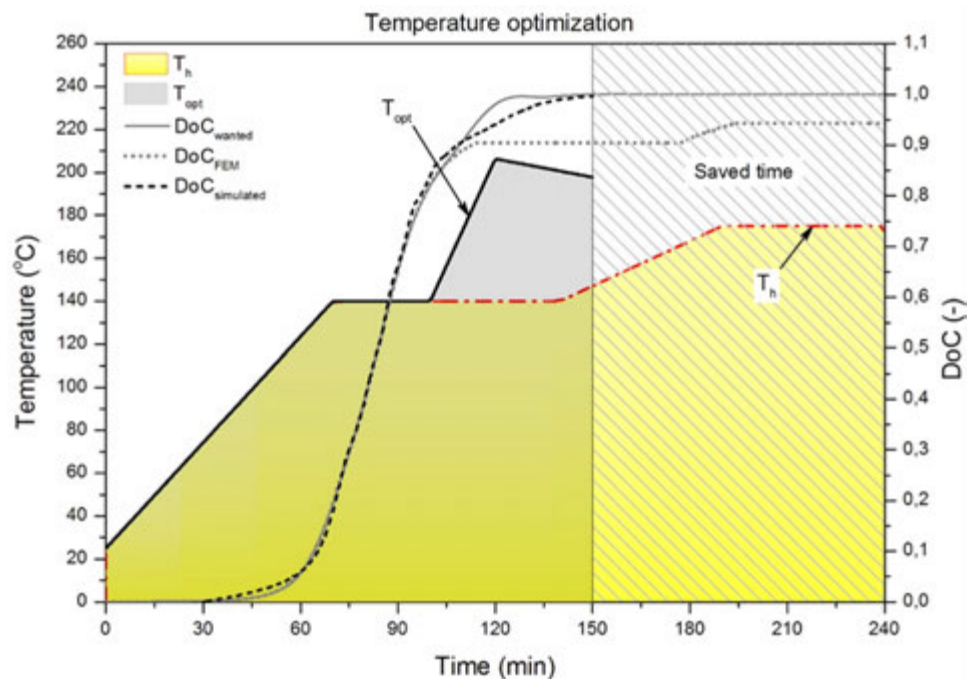


Fig. 3 Wanted DoC profile achieved by adopting the optimal autoclave thermal cycle. Reproduced from Aleksendrić, D., Carlone, P., Čirović, V., 2016. Optimization of the temperature-time curve for the curing process of thermoset matrix composites. *Applied Composite Materials* 23, 1047–1063.

implementation of neuro-fuzzy logic combinations as a decision support system has increased potentially in the span of the past decade owing to the numerous advances in comparison with its traditional counterparts.

In the next example, the ANN model reported in the previous section is coupled with a fuzzy logic controllers (FLC) to introduce the unpredictability arising from the unambiguous inputs and vague measurements of the process variables in the

manufacturing decisions making. In detail, the ANN can be employed to for prediction of curing evolution in the thermal cycle considered for the study. On the other hand, by employing the FLC, the adjustment of the thermal cycle is possible by considering and following the well-defined rules. Specifically, with FLC there is an opportunity of forming the curing cycle correction strategies by considering the factors such, temperature and DoC histories, practical constraints associated with resin carbonization temperature and DoC gradient of processing materials (Aleksendrić *et al.*, 2019). Also, in this exercise, the data necessary for the ANN models training and testing are generated using a validated FE model of the thermal curing process (Michaud *et al.*, 2002a,b; Sorrentino and Tersigni, 2012; Sorrentino *et al.*, 2015). The curing profile has been computed starting from the thermal cycle suggested by the manufacturer ("Suggested" cycle). The function of the controller will be effective if deviation occurred in the tested DoC as well as wanted profile. It also stays effective in the case of the DoC or the temperature inside the composite exceed the enforced constraints. Fig. 4 illustrates the working principle of the proposed approach along with that further details can be found in (Aleksendrić *et al.*, 2019). In the same figure, the setup used for validation is depicted. The dual ANNs models provide the prediction of the temperature and the degree-of-cure of composite using the imposed time – temperature history as an input; followed by that, the FLC compares the predicted and desired profiles and returns a set of changes to be applied to the thermal cycle. These modifications upon implementation by the ANNs serve the re-calculation of the temperature and DoC of the material. Furthermore, it is essential to reiterate the suggested steps along the whole thermal cycle.

Another added advantage of the model is that it enables the user in assessing the temperature peaks formation during the cure. The input thermal cycle data will be utilized by the ANN-DoC model to predict the DoC trend inside the composite material (Aleksendrić *et al.*, 2019). Along the composite thickness, temperature Vs time and DoC Vs time profiles were generated. Three different locations, namely the composite laminate center of and the interfaces with the top and bottom surfaces were considered. Different neural network architectures were generated to develop the model of the composite temperature, the one displaying the best prediction capability and stability was identified. It consisted of dual hidden layers having five neurons in the first and three in the second. As for the ANN-DoC model, model trained by the Bayesian Regularization algorithm returned the best prediction and generalization capabilities. The architecture consisted of again a two-layered neural model having ten neurons in the first and nine neurons in the second (Aleksendrić *et al.*, 2019).

The FLC integrates the influence of the following input variables:

- (1) Input time-temperature profile,
- (2) Difference between the maximum temperature reached in composite and carbonization temperature of the resin,

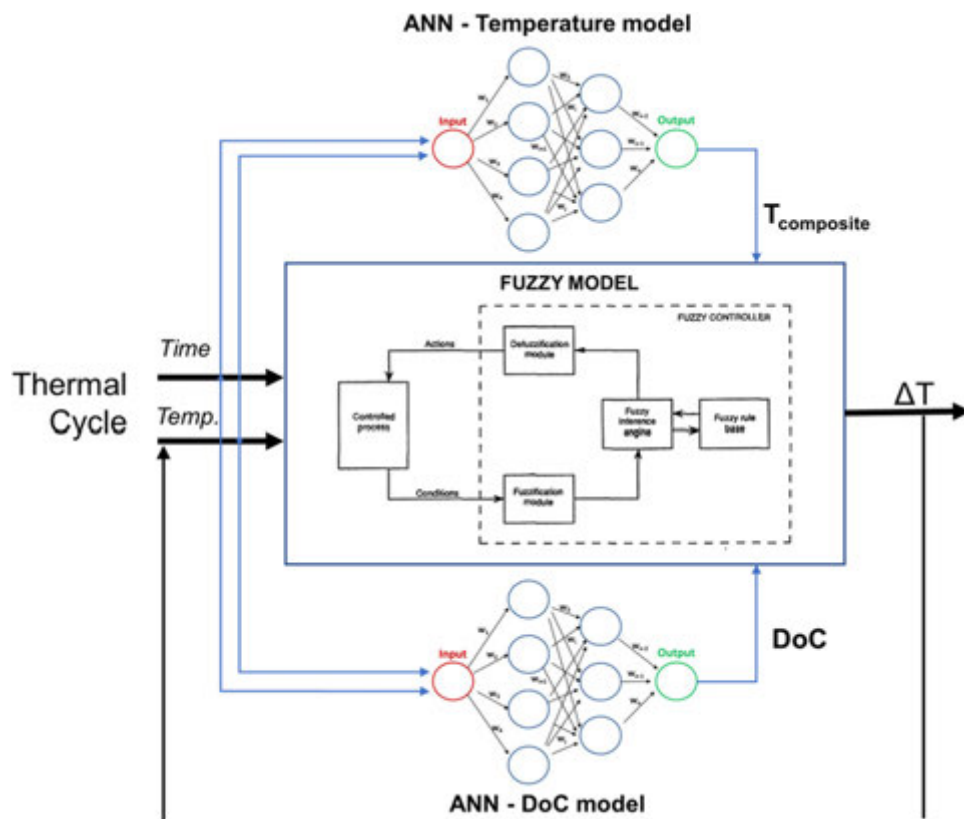


Fig. 4 Conceptual scheme of the integrated neuro-fuzzy method for resin cure optimization. Reproduced from Aleksendrić, D., Bellini, C., Carlone, P., *et al.*, 2019. Neural-fuzzy optimization of thick composites curing process. *Materials and Manufacturing Processes* 34, 262–273.

- (3) DoC difference between the predicted and desired degree-of-cure in the middle location of composite,
- (4) Difference of DoC predicted by ANN between top surface and center of a composite.

Incorporation of these input influencing factors followed by correlation with the required value for cycle temperature correction (ΔT) provided the FLC output as shown in Fig. 4. The model contains the control of the cycle temperature in order to be able to attain the wanted DoC. It occurs with the consideration of the restrictions related to the maximum temperature and gradients.

Figs. 5 and 6 provide a graphical summary of the findings of the optimization event (Aleksendrić *et al.*, 2019).

As indicated by Fig. 5, the controller for the rapid temperature enhancement after the cure process beginning. The degree of cure quickly increased at 30 min of the process consequently to the rise of the cycle temperature above 100°C. However, the imposed constraints limited the further temperature increasing needed to accelerate the polymerization. The FLC sharply reduce the heat input at 40 min, as soon the DoC gradient between the surface and center exceeded the threshold of 0.2. As a result, the temperature as well as DoC profiles are generated by the ANN-FLC model and are found to contain resemblance to that of obtained in the first scenario that probably is the representation of the optimum processing time and cost reduction which can be attainable with the apparent consideration of the posed restrictions. Hence, in this manner, it is feasible for the developed method

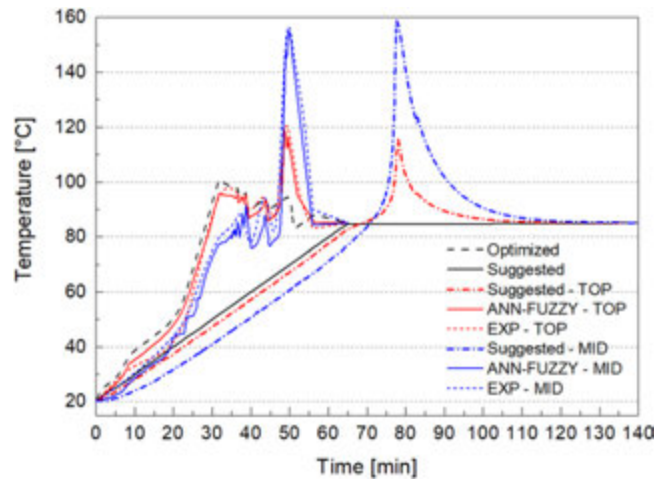


Fig. 5 Temperature profiles in the second optimization scenario. The thermal cycle suggested by the supplier ("Suggested") is depicted with solid black line. The optimized cycle is reported in black dash line. The temperature profiles calculated by FEM according to the suggested cycle, the profiles optimized by ANN-FUZZY control as well as the measured trends (EXP) are depicted with dash-dot, solid and dash blue and red lines, respectively. Reproduced from Aleksendrić, D., Bellini, C., Carlone, P., *et al.*, 2019. Neural-fuzzy optimization of thick composites curing process. Materials and Manufacturing Processes 34, 262–273.

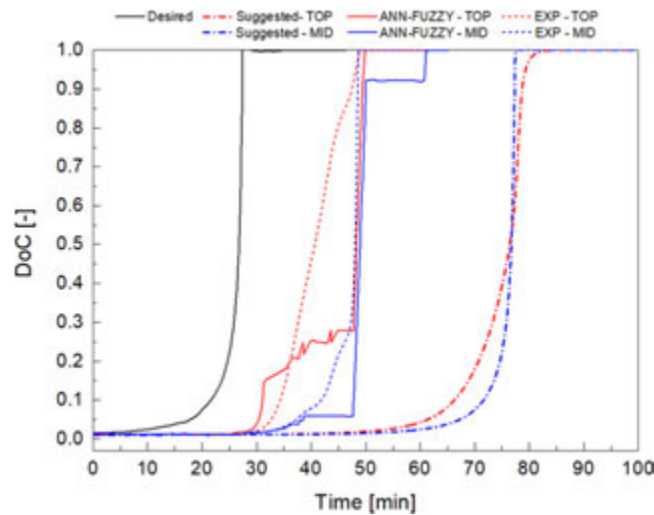


Fig. 6 Second optimization scenario: DoC profile evaluated on top and center locations. The desired profile, FEM outcomes, ANN-FUZZY predictions and the experimentally profiles were compared. Reproduced from Aleksendrić, D., Bellini, C., Carlone, P., *et al.*, 2019. Neural-fuzzy optimization of thick composites curing process. Materials and Manufacturing Processes 34, 262–273.

to attend to the exaggerated requests and offer a proper thermal cycle that will take into account the process restrictions. The differences found among the experimental and ANN-FLC trends were admissible for the temperature profiles. Despite that, the remarkable divergence was observed for the DoC profiles, specifically on the top location where experiments and predictions display an obvious mismatch (Aleksendrić *et al.*, 2019).

Conclusions

In this article, some results provided by neural-based optimization of the curing process is discussed. ANN models, predicting the temperature and DoC profiles, were coupled with genetic algorithms and fuzzy logic controllers to design the optimal curing stage of the considered composites with a certain thickness. The following remarks are outlined:

- (1) Autoclave temperature influences the DoC change at a particular time instance. The neuro-genetic optimization model was able to recognize this influence of autoclave temperature optimization when the thermal cycle was imposed. Specifically, in the said thermal cycle a dynamic adaptation of the autoclave temperature can result in the maximization and rapid growth of the DoC. This approach offers an opportunity of flexible customization of the autoclave curing process concerning the preference of the desired degree of cure and its gradient of change against various thermal influences (minute, normal or rapid heating).
- (2) The neural-fuzzy controller presents the ability involving the prompt modification of the input cycle temperature in cases when the process control variables, i.e. a composite degree of cure is found to differ from the desired values or when the imposed constraints are not fulfilled. This approach contains the possibility of achieving a significant reduction in the curing process duration in comparison with that of the supplier thermal cycle.
- (3) Useful development should be reasonably directed toward the extension of the model, in order to include also stress-strain computation and the implementation of the neural-fuzzy controller in on-line sensor-based monitoring and control systems.

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3D Topology Optimization of Continuous Fiber-Reinforced Structures

Alexander A Safonov, Skolkovo Institute of Science and Technology, Moscow, Russia

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Introduction

In the last years the industry has seen a rapid development in the application of 3D printing technologies for fabrication of fiber-reinforced plastic structures (Zhang *et al.*, 2016). The application of the modified fused-deposition modeling (FDM) process (Parandoush and Lin, 2017; Iragi *et al.*, 2019) to fabricate thermoplastic structures with continuous fiber reinforcement (CFR) is a good example of that trend. In the FDM process, a thermoplastic filament and a continuous fiber strand are separately fed into a 3D printer (see Fig. 1(a)). The filament is then heated inside a heated nozzle to a melting point and melted thermoplastic impregnates a reinforcing fiber immediately before deposition (Matsuzaki *et al.*, 2016). As compared to traditional 3D printed composites (Ahmed *et al.*, 2020), printed CFR composites can offer an order of magnitude better performance (Melenka *et al.*, 2016). A digital model of a structure is translated into a set of commands for a 3D printer by the custom software plugin that defines the layout of reinforced and non-reinforced plastic, and specifies the orientation of reinforcement. Fig. 1 shows the printed continuous fiber-reinforced 3D parts manufactured by the Anisoprint process (Azarov *et al.*, 2017; Adumitroaie *et al.*, 2019; Azarov *et al.*, 2019). The process of 3d printing of composites was tested earlier in prototyping various components, such as unmanned aerial vehicle composite frame structure (Azarov *et al.*, 2019), locally resonant carbon-fiber composite meta-structures for attenuation of broadband vibration (Mizukami *et al.*, 2021), bolted joints (Sugiyama *et al.*, 2020) etc. The use of industrial robots can significantly extend 3D printing capabilities, making it possible to produce large composite parts with complex spatial architecture and higher fiber content (Zhang *et al.*, 2016). However, to take the full advantage of these capabilities engineers should use optimized design methods. This will require the development of structural optimization methods adapted for composite structures, making it possible to achieve best mechanical performance with minimum weight of a structure. This implies a capability to find density distribution and local orientation of reinforcement in 3D composite structures.

Currently, spatial optimization of structures is the subject of several studies (Bendsoe, 2003; Aage *et al.*, 2017). Standard topology optimization methods consist in modeling the layout of material using the material density parameter, ρ , varying from 0 to 1, meaning respectively the absence or presence of material. To describe the relationship between structural properties and the density of material a power law is used. The method describing the relationship between mechanical properties and the density is known as Solid Isotropic Material with Penalization (SIMP) (Bendsoe, 2003; Bendsoe, 1989). Thus, we reduce the problem of optimization to finding an optimum distribution of ρ : $\min_f(\rho)$. However, the application of non-gradient methods for topology optimization of models consisting of more than 1000 elements, which is common in engineering practice, becomes extremely expensive task (Rozvany, 2009). Among gradient-based methods of topology optimization (Sigmund and Maute, 2013; Svanberg, 1987; Ostanin *et al.*, 2017; Ai and Gao, 2019) special attention should be given to those reducing a topology optimization problem to a problem of finding a stationary point of an Ordinary Differential Equation (ODE) (Klarbring and Torstenfelt, 2010). Taking into account density constraints, ρ , the right term of an ODE will be equal to a projected negative gradient of the objective function. The explicit Euler algorithm can be used to find numerical schemes of topology optimization solution. As we can see in (Klarbring and Torstenfelt, 2012), iterative schemes match the algorithms used in bone remodeling studies (Harrigan and Hamilton, 1994). Also shown is a connection between optimization algorithms and natural phenomena, such as amenorrhea organism growing toward food sources (Safonov and Jones, 2017; Jones and Safonov, 2018), and formation of natural sandstone arches (Ostanin *et al.*, 2017; Safonov, 2018; Safonov *et al.*, 2020).

Clearly, the obvious approach to simultaneous optimization of density and orientation distribution for 2D structures will consist in aligning reinforcement along the principal stress direction (fiber steering technique) (Ghiasi *et al.*, 2010) and using traditional topology optimization tools to find material distribution. The basis for this approach is the fact that extreme energy values in case of a planar problem with orthotropic material are achieved with coinciding principal directions for material, stresses and strains (Pedersen, 1989, 1990). Although, there may exist optimal solutions where coinciding directions of principal stresses and strains are not aligned in the direction of principal axes of a material. In (Pedersen, 1990; Banichuk, 1979) it was shown that for materials weak in shear the optimum orientation of reinforcement and the direction of principal stress would coincide. Besides, for a 3D setting, as was found in (Rovati and Taliercio, 2003; Norris, 2006), directions of principal stresses and strains will coincide for critical values of strain energy density. For the transversely isotropic material, Norris (2006) has established a constraint for the elastic constants, such that the global energy minimum can only occur when the axis of anisotropy is aligned with one of the principal stress axes (Norris, 2006). Cheng *et al.* (1994), have demonstrated that the principal stress method (Suzuki and Kikuchi, 1991) produces results that are very close to those yielded by the general stress-based optimality criteria method (Hassani and Hinton, 1998). Setoodeh *et al.* (2005) proposed a method to design a topology and fiber path in composite laminae simultaneously, using a cellular automata (CA) framework. In (Nomura *et al.*, 2015; Lee *et al.*, 2018) the general topology optimization method based on isoparametric projection have been proposed by Nomura, Lee *et al.* for continuous and discrete design problems.

In spite of a relatively small number of works on combined design of orientation and topology (Setoodeh *et al.*, 2005; Tong *et al.*, 2017), their authors have succeeded in solving several interesting problems for composite 2D structures, such as the plate with a centrally located circular hole (Hyer and Charette, 1991; Pedersen, 1991), cantilever plates (Setoodeh *et al.*, 2005; Lee *et al.*, 2018; Pedersen, 1991; Desai *et al.*, 2021; Kim *et al.*, 2020), membrane structures (Klarbring *et al.*, 2017), compliant mechanisms (Tong *et al.*, 2017).

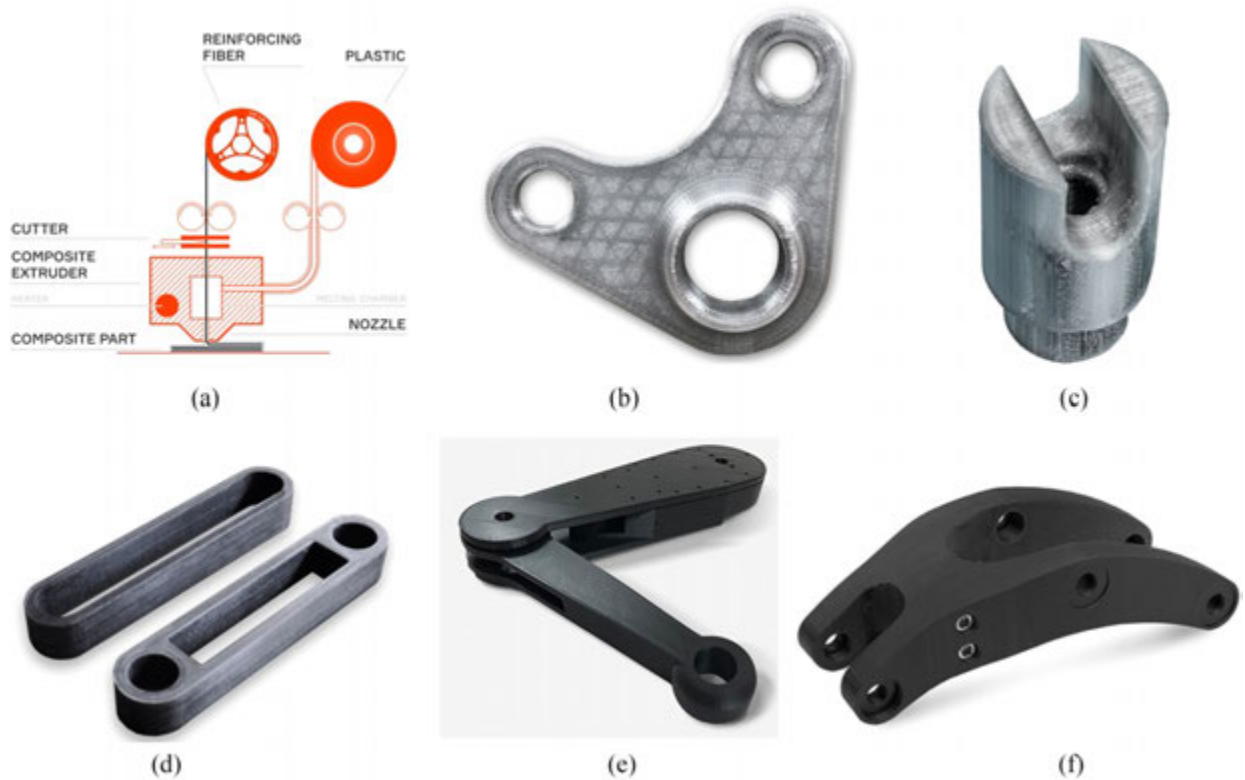


Fig. 1 Fabrication of continuous fiber reinforced 3D printed parts by Anisoprint process: (a) – process scheme; (b) - wall bracket; (c) - clevis for production line of dairy brand; (d) - filament winding machine roller mount; (e) - legs of mobile robot for sensing, inspection, and remote operation; (f) - Scott Gambler downhill bike composite rocker. Reproduced from Azarov, A.V., Antonov, F.K., Vasil'ev, V.V., *et al.*, 2017. Development of a two-matrix composite material fabricated by 3D printing. *Polym. Sci. Ser. D* 10, 87–90. <https://doi.org/10.1134/S1995421217010026>. Adumitroaie, A., Antonov, F., Khaziev, A., *et al.*, 2019. Novel continuous fiber bi-matrix composite 3-D printing technology. *Materials* 12 (11), 3011. <https://doi.org/10.3390/ma12183011>. Azarov, A.V., Antonov, F.K., Golubev, M.V., Khaziev, A.R., Ushanov, S.A., 2019. Composite 3D printing for the small size unmanned aerial vehicle structure. *Compos. Part B Eng.* 169, 157–163. <https://doi.org/10.1016>.

The studies discussed above deal with optimization of reinforcement lay-up in 2D objects, such as plates or membranes. Only a few studies over the last two years have dealt with 3D structures (Safonov, 2019; Yan *et al.*, 2020; Caivano *et al.*, 2020; Nosouhi Dehnavi *et al.*, 2020; Schmidt *et al.*, 2020). However, the optimization of 3D printed composite structures will require new topology optimization methods. Such methods should be tailored for structures with continuous fiber reinforcement, offer extensive engineering capabilities, high computing speed, and be easy to learn. In this study authors propose the new approach to finding the distribution of density and the vector of fiber orientation in composite 3D structures made of transversely isotropic materials. The method uses a dynamical systems approach to find density distribution (Klarbring and Torstenfelt, 2010; Klarbring and Torstenfelt, 2012), in combination with the method of orienting the reinforcement in the direction of principal stress with minimum compliance (Suzuki and Kikuchi, 1991). In (Safonov, 2019) the evolutionary algorithm is discussed, where the search for material density distribution and for local vector of reinforcement, $\mathbf{n}$, is performed simultaneously. In order to simplify the algorithm, in this study we propose the two stage approach. The first stage is the search for optimal density distribution, assuming the material to be isotropic; and at the second stage we find the distribution of the local vector of reinforcement, $\mathbf{n}$, for a given density distribution for transversely isotropic material being studied. The proposed optimization algorithm is implemented in the ABAQUS finite element analysis suite via the user subroutines mechanism. The subroutines used in the study are available in the GitHub open access repository. The optimal distribution of material density and fiber orientation vector are determined for three structural cases used as benchmarks: 2D beam, 3D cube, and 3D cantilever beam. The results are compared with those obtained by Setoodeh *et al.* (2005) and Safonov (2019).

Method

A classical topology optimization problem consists in finding the optimum layout of material to ensure maximum stiffness of a structure (Bendsoe, 2003). In other words, topology optimization deals with finding an optimum distribution of material density

ρ and a local reinforcement (fiber orientation) vector $\mathbf{n}$ within a given volume Ω , such that the compliance of structure is minimum (Sigmund, 2001):

$$\text{Minimize } C(\rho, \mathbf{n}) = \frac{1}{2} \int_{\Omega} (\varepsilon_{xyz})^T \cdot \mathbf{E}^*(\rho, \mathbf{n}) \cdot \{\varepsilon_{xyz}\} d\Omega \quad (1)$$

where

$$\mathbf{E}^*(\rho, \mathbf{n}) = \rho^p \mathbf{E}(\mathbf{n}), p > 1. \quad (2)$$

The following constraints are imposed on the distribution of density, ρ :

$$\int_{\Omega} \rho d\Omega \leq M; 0 < \rho_{\min} \leq \rho \leq 1. \quad (3)$$

The following notation is used in equations above: $\{\varepsilon_{xyz}\}$ - strains in Voigt notation, $\mathbf{E}^*(\rho, \mathbf{n})$ - transformation of local stiffness matrix in xyz -coordinates, $\mathbf{E}(\mathbf{n})$ - transformation of local stiffness matrix at $\rho(x) = 1$, M - constraint on the amount of material used, p - penalization factor ($p > 1$), $\rho_{\min}$ - minimum density. We assume the composite material to be transversely isotropic and the axis of anisotropy to be aligned with the local vector of reinforcement $\mathbf{n}$. Then we express $\mathbf{E}(\mathbf{n})$, using the stiffness matrix of transversely isotropic material in the principal coordinate system (123-coordinates) $[\mathbf{Q}]$, which is the inverse of the compliance matrix $[\mathbf{S}]$ Akkerman (2002),

$$[\mathbf{E}(\mathbf{n})] = [\mathbf{T}] \cdot [\mathbf{Q}] \cdot [\mathbf{R}] \cdot [\mathbf{T}]^{-1} \cdot [\mathbf{R}]^{-1}, \quad (4)$$

where $[\mathbf{Q}] = [\mathbf{S}]^{-1}$, and $[\mathbf{S}]$ is the compliance matrix in principal coordinate system,

$$[\mathbf{S}] = \begin{bmatrix} 1/E_1 & -\nu_{12}/E_1 & -\nu_{12}/E_1 & 0 & 0 & 0 \\ -\nu_{12}/E_1 & 1/E_2 & -\nu_{23}/E_2 & 0 & 0 & 0 \\ -\nu_{12}/E_1 & -\nu_{23}/E_2 & 1/E_2 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1/G_{23} & 0 & 0 \\ 0 & 0 & 0 & 0 & 1/G_{12} & 0 \\ 0 & 0 & 0 & 0 & 0 & 1/G_{12} \end{bmatrix}, \quad (5)$$

$[\mathbf{R}]$ is Reuter's matrix,

$$[\mathbf{R}] = \begin{bmatrix} 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 2 & 0 & 0 \\ 0 & 0 & 0 & 0 & 2 & 0 \\ 0 & 0 & 0 & 0 & 0 & 2 \end{bmatrix} \quad (6)$$

$[\mathbf{T}]$ is the transformation matrix (Newnham, 2005),

$$[\mathbf{T}] = \begin{bmatrix} a_{11}^2 & a_{12}^2 & a_{13}^2 & 2a_{12}a_{13} & 2a_{13}a_{11} & 2a_{11}a_{12} \\ a_{21}^2 & a_{22}^2 & a_{23}^2 & 2a_{22}a_{23} & 2a_{23}a_{21} & 2a_{21}a_{22} \\ a_{31}^2 & a_{32}^2 & a_{33}^2 & 2a_{32}a_{33} & 2a_{33}a_{31} & 2a_{31}a_{32} \\ a_{21}a_{31} & a_{22}a_{32} & a_{23}a_{33} & a_{22}a_{33} + a_{23}a_{32} & a_{21}a_{33} + a_{23}a_{31} & a_{22}a_{31} + a_{21}a_{32} \\ a_{31}a_{11} & a_{32}a_{12} & a_{33}a_{13} & a_{12}a_{33} + a_{13}a_{32} & a_{13}a_{31} + a_{11}a_{33} & a_{11}a_{32} + a_{12}a_{31} \\ a_{11}a_{21} & a_{12}a_{22} & a_{13}a_{23} & a_{12}a_{23} + a_{13}a_{22} & a_{13}a_{21} + a_{11}a_{23} & a_{11}a_{22} + a_{12}a_{21} \end{bmatrix}, \quad (7)$$

(a_{11}, a_{21}, a_{31}) coincide with the components of vector $\mathbf{n}$, (a_{12}, a_{22}, a_{32}) and (a_{13}, a_{23}, a_{33}) coincide with vectors appending to orthonormal basis.

Following are the iterative algorithms used to find density ρ and a local vector of reinforcement $\mathbf{n}$. Contrary to the study by Safonov (2019) where the evolutionary algorithm for simultaneous search of density distribution and local vector of reinforcement is discussed, in this work we propose the 2-stage algorithm. At the first stage we find the optimal density distribution, assuming the material to be isotropic, then, at the second stage we find the distribution of local vector of reinforcement, $\mathbf{n}$, for the transversely isotropic material at a given density distribution.

Density

In line with the SIMP method, we divide the domain being studied into finite elements with varying material density values assigned to each element (Bendsoe, 2003). Here, the material density, ρ_i , is determined for each integration point of each element. To solve the problem Eqs. (1)–(7) author uses the approach applied in dynamical systems modeling (Klarbring and Torstenfelt, 2010).

In accordance with the proposed algorithm, we find density distribution for the isotropic material, i.e. the material with compliance matrix where elastic moduli and Poisson coefficients are equal ($E_1 = E_2 = E$ and $\nu_{12} = \nu_{23} = \nu$), and shear moduli are expressed as follows $G_{12} = G_{23} = E/(2 + 2\nu)$.

Assuming that ρ depends on a time-like variable t , and considering the following differential equation to determine density, ρ_i , in i -th integration point when solving the problem stated in Eqs. (1)–(3):

$$\dot{\rho}_i = \lambda \left(\frac{pC_i(\rho_i)}{\rho_i} - \mu V_i \right), C_i(\rho_i) = \frac{1}{2} \int_{\Omega_i} \{ \varepsilon_{xyz} \}^T \cdot E^*(\rho) \cdot \{ \varepsilon_{xyz} \} d\Omega, \quad (8)$$

where dot above denotes the derivative with respect to t ; Ω_i is a domain of i -th integration point; V_i is a volume of a domain of i -th integration point; λ is a positive dimensional physical constant; μ is a positive parameter regulating the relative importance of the cost function (1) and of the mass constraint (3). To obtain this equation we can use methods of projected dynamical systems (Nagurney, 1996), or the bone remodeling method (Harrigan and Hamilton, 1994; Huijskes *et al.*, 2000; Payten *et al.*, 1998). The detailed description of relations between the Optimality Criteria (OC) method (Bendsøe, 2003), the method of dynamical systems, and the bone remodeling method was given by Klarbring in (Klarbring and Torstenfelt, 2012). The parameter μ is selected during the calculations so as to satisfy the mass constraint (3) and is usually found by using the bi-sectioning algorithm (Sigmund, 2001). In all calculations below, the value of μ at each iteration step is determined to the fourth decimal place.

Authors use a projected Euler method (Pedersen, 1991) to obtain a numerical solution of Eq. (5), giving an iterative formulation for ρ_i (Harrigan and Hamilton (1994):

$$\rho_i^{n+1} = \rho_i^n + q \left[\frac{pC_i(\rho_i^n)}{\rho_i^n V_i} - \mu^n \right], \quad (9)$$

where $q = \lambda \Delta t$, ρ_i^{n+1} and ρ_i^n – numerical approximations of $\rho_i(t + \Delta t)$ and $\rho_i(t)$.

To avoid using the trial and error method to find the dimensional parameter q in Eq. (9), we shall use a modified algorithm based on dimensionless parameters. As multiplication of the right term of Eq. (8) by a positive function does not affect the optimization outcome (Klarbring and Torstenfelt, 2010), the following modification may be considered:

$$\rho_i^{n+1} = \rho_i^n + \Delta \rho_i^n, \quad (10)$$

where $\rho_i^0 = \rho_0$ – specified initial value of density, and $\Delta \rho_i^n$ – is determined as follows

$$\Delta \rho_i^n = \begin{cases} K \Delta \rho_i^{n-1} \text{ if } \left(\frac{pC_i(\rho_i^n)}{\rho_i^n V_i} - \mu^n \right) \Delta \rho_i^{n-1} > 0, \\ -\Delta \rho_i^{n-1} / K \text{ if } \left(\frac{pC_i(\rho_i^n)}{\rho_i^n V_i} - \mu^n \right) \Delta \rho_i^{n-1} \leq 0, \end{cases} \quad (11)$$

where $\Delta \rho_i^0 = k_0$; K , k_0 – positive constants. With matching signs of equation $\left(\frac{pC_i(\rho_i^n)}{\rho_i^n V_i} - \mu^n \right)$ and the previous increment of $\Delta \rho_i^{n-1}$, the current increment $\Delta \rho_i^n$ will increase K times ($K \geq 1$), compared to $\Delta \rho_i^{n-1}$; therefore, introduction of K parameter will result in accelerated convergence rate of the algorithm. Otherwise, the increment $\Delta \rho_i^n$ will decrease K times and reverse its sign.

After a value of ρ_i^{n+1} is calculated using Eq. (12), we make a projection onto a set of constraints for ρ_i :

$$\rho_i^{n+1} = \begin{cases} 1 \text{ if } \rho_i^{n+1} > 1 \\ \rho_i^{n+1} \text{ if } \rho_{\min} \leq \rho_i^{n+1} \leq 1 \\ \rho_{\min} \text{ if } \rho_{\min} > \rho_i^{n+1} \end{cases} \quad (12)$$

Discretization approach considered here may yield numerical instabilities such as mesh dependence and checkerboard patterns (Sigmund and Petersson, 1998). To prevent possible problems, we apply the density filtering technique (Sigmund, 2001); the filter replaces density values ρ_i within all modeling domain with filtered density value when computing mechanical characteristics (2). The filter smooths the density values over adjacent integration points and is implemented as follows (Klarbring and Torstenfelt, 2010):

$$\bar{\rho}_j = \sum_{i=1}^E \Psi_{ji} \rho_i, j = 1, \dots, E. \quad (13)$$

Here

$$\Psi_{ij} = \frac{\psi_{ij} V_j}{\sum_{k=1}^E \psi_{ik} V_k}, \psi_{ij} = \max \left(0, 1 - \left| \mathbf{e}_i - \mathbf{e}_j \right| \frac{R}{R} \right), \quad (14)$$

where $\mathbf{e}_i$ is the position vector of the integration point i , R is the filter radius, and V_j – the volume of the j -th integration point.

Local Vector of Reinforcement

In (Norris, 2006) the constraint for the elastic constants of transversely isotropic material is obtained, such that the global energy minimum can only be reached when the axis of anisotropy is aligned with one of the principal stress axes. The constraint consists

Table 1 Algorithm parameters used in simulation

ρ	R	K	k_0	ρ_{min}
3	1.42	1.2	0.01	0.01

in negative value of the parameter $a = S_{11} + S_{33} - 2S_{13} - 4S_{55}$, where S_{11} , S_{33} , S_{13} , S_{55} – elements of the compliance matrix $[S]$ (5). For the transversely isotropic material studied further (see [Table 1](#)) this condition is satisfied: $a = -0.517 \text{ GPa}^{-1} < 0$. Here we consider the algorithm that aligns the local direction of reinforcement in such a way as to minimize compliance of 3D structures. The iterative formulation used to find reinforcement direction is expressed as assigning the current position of reinforcement axis $\mathbf{n}_i^n$ in i -th integration point toward the selected direction of principal stress, in the following form:

$$\mathbf{n}_i^{n+1} = \mathbf{n}_p^n i, \quad (15)$$

where $\mathbf{n}_p^n i$ – the vector of chosen direction of principal stress $\mathbf{n}_p^n i$. The principal direction is selected using the following expression:

$$\mathbf{n}_p^n i = \operatorname{argmin}_{\mathbf{n}_x^n i} \left(\frac{1}{2} \{ \sigma_{xyz} \}^T \cdot [\mathbf{E}^*(\rho_i^n, \mathbf{n}_x^n i)]^{-1} \cdot \{ \sigma_{xyz} \} \right), \alpha = 1, 2, 3, \quad (16)$$

where $\mathbf{n}_x^n i$ – the principal stress vector; $\mathbf{E}^*(\rho_i^n, \mathbf{n}_x^n i)$ – the transformed stiffness matrix where a local vector of reinforcement is coincident with $\mathbf{n}_x^n i$; $\{ \sigma_{xyz} \}$ – the stresses. The expression [Eq. \(16\)](#) includes the principal stress vector $\mathbf{n}_x^n i$ such that the local compliance for local stresses $\{ \sigma_{xyz} \}$ for transformed stiffness matrix $\mathbf{E}^*(\rho_i^n, \mathbf{n}_x^n i)$ is minimal.

In order to estimate the number of independent design variables used to find the density [Eqs. \(12\)–\(14\)](#) and the direction of reinforcement [Eqs. \(15\), \(16\)](#) we can multiply the number of elements in a model, the number of integration points in the element of a given type, and the number of design variables for each integration point (the density and 2 independent design variables for the direction of reinforcement in the 3D setting). Thus, the number of independent design variables for a 3D model of 10,000 8-node linear brick elements will constitute 240,000.

Implementation Within Abaqus Environment

The algorithm shown above is implemented within Abaqus environment with user subroutines Uexternaldb, Urdfil, Usdflid ([Abaqus Analysis User Manual, 2014](#)). We use Uexternaldb subroutine to set specific parameters of the algorithm, Urdfil — to access the results of stress-strain distribution computations by Abaqus Solver, and Usdflid procedure — to account for changes in mechanical properties. In order to determine parameters of the local vector of reinforcement $\mathbf{n}_i^n$ we use the Usdflid procedure and the spherical coordinate system:

$$\mathbf{n}_i^n = (\cos\theta, \sin\theta\cos\varphi, \sin\theta\sin\varphi), \quad (17)$$

where θ – polar angle ($0^\circ \leq \theta \leq 90^\circ$) and φ – azimuth angle ($-180^\circ \leq \varphi \leq 180^\circ$). The uniform grid with 10° increments was used to partition values of θ and φ angles.

[Fig. 2](#) shows the diagram of optimization process implemented in ABAQUS. Subroutines call locations are marked with red. The subroutines used in the study are available in the GitHub open access repository ([Safonov, 2020](#)).

It is worth noting that the developed plug-in makes it possible to optimize computational models built with different elements types with different number of integration points, while taking into account contact interactions and geometric nonlinearity. The plug-in also allows users to optimize the density distribution or the reinforcement orientation separately, and to isolate certain parts of a model that do not need to be optimized. Moreover, users can assign different filtering radii for density distribution and reinforcement orientation optimizations, allowing for variations in typical dimensions of structural elements and for reinforcement orientation variability.

Although we used a uniform rectangular mesh of the same type elements in the test problems, the proposed method and developed plug-in make it possible to optimize an arbitrary mesh of finite elements.

Results and Discussion

Comparison With the Previous Research Data

In this section the results obtained with proposed method are presented and compared with those reported by [Setoodeh et al. \(2005\)](#) and [Safonov \(2019\)](#). For the purposes of this study we used the mechanical properties data presented in ([Setoodeh et al., 2005](#)). The algorithm parameters are given in [Table 1](#). In this study we consider two predictive models: the symmetric cantilever plate with aspect ratio of 4:1 (325×82 finite elements) and the asymmetric cantilever plate with aspect ratio of 2:1 (203×102 finite elements). The initial position of the axis of anisotropy is coincident with OX axis for all finite elements. [Table 2](#) shows the normalized compliance values with respect to a 0° fiber design with initial mean density of $\rho_{ev}^0 = 1.0$ for reviewed models with various target values of mean density $\rho_{ev}^\infty = M/V$ (volume fraction) for the proposed method, and the results reported in ([Setoodeh et al., 2005](#)) and ([Safonov, 2019](#)). [Fig. 3](#) shows the topology of the optimal designs along with the fiber orientation angles for symmetric cantilever plate. [Fig. 4](#) shows the topology of the optimal designs along with the fiber orientation angles for asymmetric cantilever plate.

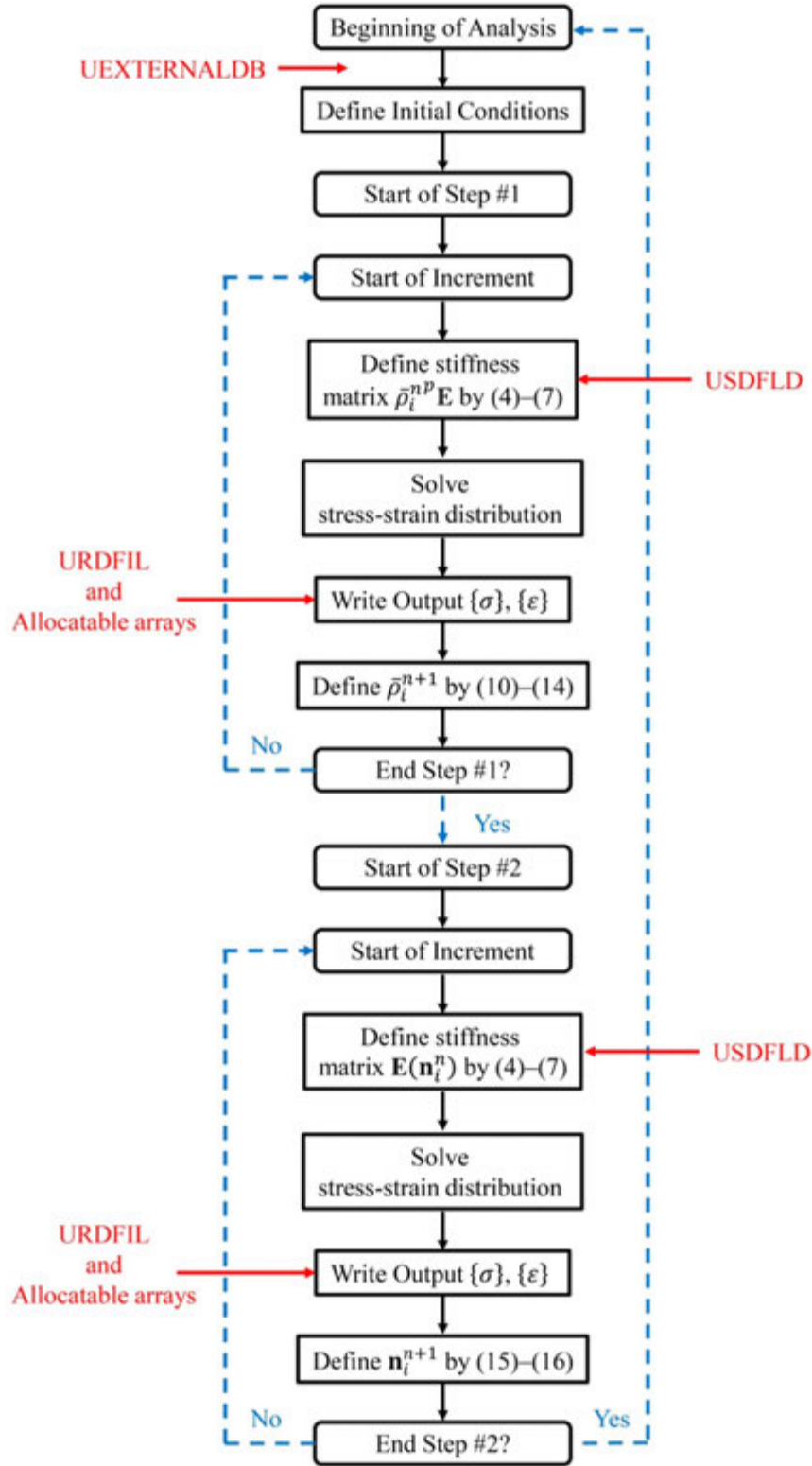


Fig. 2 The diagram of optimization process implemented in Abaqus.

For all the considered cases, the proposed method shows better results than those obtained in [Safonov \(2019\)](#). For obtained results by [Setoodeh et al. \(2005\)](#), the proposed method produced better results in case of the symmetric cantilever plate. Moreover, at the volume fraction of $\rho_{ev}^{\infty} = 0.3$ the structure obtained with proposed method demonstrated the 29% lower compliance, compared to that obtained by [Setoodeh et al. \(2005\)](#). However, at the volume fraction of $\rho_{ev}^{\infty} = 0.5$ for the asymmetric cantilever

Table 2 Normalized compliance values for cantilever plates, obtained using the proposed method, and borrowed from Setoodeh et al. and Safonov

Volume fraction (target mean density ρ_{ev}^∞)	Normalized compliance		
	Proposed Method	Setoodeh (Hassani and Hinton, 1998)	Safonov (Safonov, 2019)
Symmetric cantilever plate (325×82 finite elements)			
0.7	0.86	0.88	–
0.5	1.05	1.14	1.12
0.3	1.57	2.22	1.86
Asymmetric cantilever plate (203×102 finite elements)			
0.7	0.42	0.36	–
0.5	0.52	0.47	0.53

Note: Setoodeh, S., Abdalla, M.M., Gürdal, Z., 2005. Combined topology and fiber path design of composite layers using cellular automata. *Struct. Multidiscip. Optim* 30, 413–421. Available at: <https://doi.org/10.1007/s00158-005-0528-y>. Safonov, A.A., 2019. 3D topology optimization of continuous fiber-reinforced structures via natural evolution method. *Compos. Struct.* 215, 289–297. Available at: <https://doi.org/10.1016/j.compstruct.2019.02.063>.

plate the proposed method resulted in a less stiffer structure with 11% higher compliance compared to that reported by Setoodeh et al. (2005). Unfortunately, Setoodeh et al. (2005) did not specify the load application technique, which is essential for the problems studied here. This is particularly true for the case of asymmetric cantilever plate. In this case a substantial vertical load is applied to the corner region, creating the zone of considerable stress concentration. Also, Setoodeh's work (Setoodeh et al., 2005) lacks the data on absolute compliance values for the initial 0° fiber design with initial mean density of $\rho_{ev}^0 = 1.0$, and for obtained optimized structures. The absence of these data makes it impossible to validate the models described here against those used by Setoodeh et al. (2005). It should be noted that other studies mentioned in the Introduction section also lack the data necessary for the full scale comparison of reported optimization results with structures obtained with proposed algorithm.

Sample Problems

Following is the detailed description of computations conducted for the tested 2D and 3D structures. Three test problems were used to test the developed algorithm: the bending of simply supported 2D beam carrying central concentrated load; the loading of 3D cube by concentrated vertical load; and the bending of 3D cantilever beam (see Fig. 5).

The first problem is the optimization of a flat rectangular beam carrying concentrated load $F = -200\text{kN}$. The load is applied to the center of the upper side of the beam and acts in downward direction. As only half of the model is considered (see Fig. 5) we apply the vertical load of $F/2$ to the upper left corner of the model. The load is uniformly distributed between 2 nodes of the element adjacent to the upper left corner of the model. Zero vertical displacement conditions are imposed on the lower right vertex. The model has the length of $a = 150\text{m}$, and the height of $b = 50\text{m}$. To model the design domain we use a mesh of $150 \times 50 \times 1$ cubic C3D8-type elements with 8 integration points. Final compliance value for target mean density of ρ_{ev}^∞ is set equal to 0.3. Thus, the number of design variables used to find density distribution alone will constitute 60 000. The simultaneous search for density distribution and reinforcement direction will increase the number of involved design variables to 180 000.

The second problem deals with optimization of a 3D cube carrying concentrated vertical load of $F = -900\text{kN}$. The load is applied to the center of the upper face and is uniformly distributed between 9 nodes of 4 finite elements adjacent to the center of the of the upper face and located on the upper face (see Fig. 5). The model is rigidly fixed at its lower face vertices, and constraints are imposed on the square pads adjacent to the vertices. Dimensions of pads are $2 \times 2\text{m}$. The edges of the cube have length of $c = 20\text{m}$. To model the design domain we use the mesh of $20 \times 20 \times 20$ of C3D8-type cubic elements with 8 integration points. The final compliance value for target mean density of ρ_{ev}^∞ is set equal to 0.1. Thus, the number of design variables used in finding density distribution will constitute 64,000. The simultaneous search of density distribution and reinforcement orientation will increase this number to 192,000.

The third problem deals with optimization of the 3D cantilever beam carrying the vertical concentrated load of $F = -200\text{kN}$. The load is applied to the center of the extreme lower horizontal face and acts in downward direction. Only half of the model is considered, with zero displacement conditions imposed on the opposite vertical faces. The model has the length of $d = 40\text{m}$, the height of $e = 20\text{m}$, and the width of $f = 10\text{m}$. To model the design domain we use the mesh of $40 \times 20 \times 10$ cubic C3D8-type elements with 8 integration points. Final compliance value for target mean density ρ_{ev}^∞ is set equal to 0.1. Thus, the number of design variables involved in finding density distribution alone will constitute 64,000. The simultaneous search for density distribution and reinforcement direction will increase the number of involved design variables to 192,000.

Following materials were used in simulations: unidirectionally reinforced material (UD), quasi-isotropic laminate (QIL), and 3D quasi-isotropic bulk material (QIB). The quasi-isotropic laminate (QIL) is built with UD material (Akkerman, 2002). The 3D QIB material represents an artificial isotropic material built with UD material. Mechanical properties of materials are given in Table 3. Properties of UD and QIL are taken from (Akkerman, 2002). Properties of QIB material are proposed by the author of this study. Algorithm parameters used in solving test problems are given in Table 1.

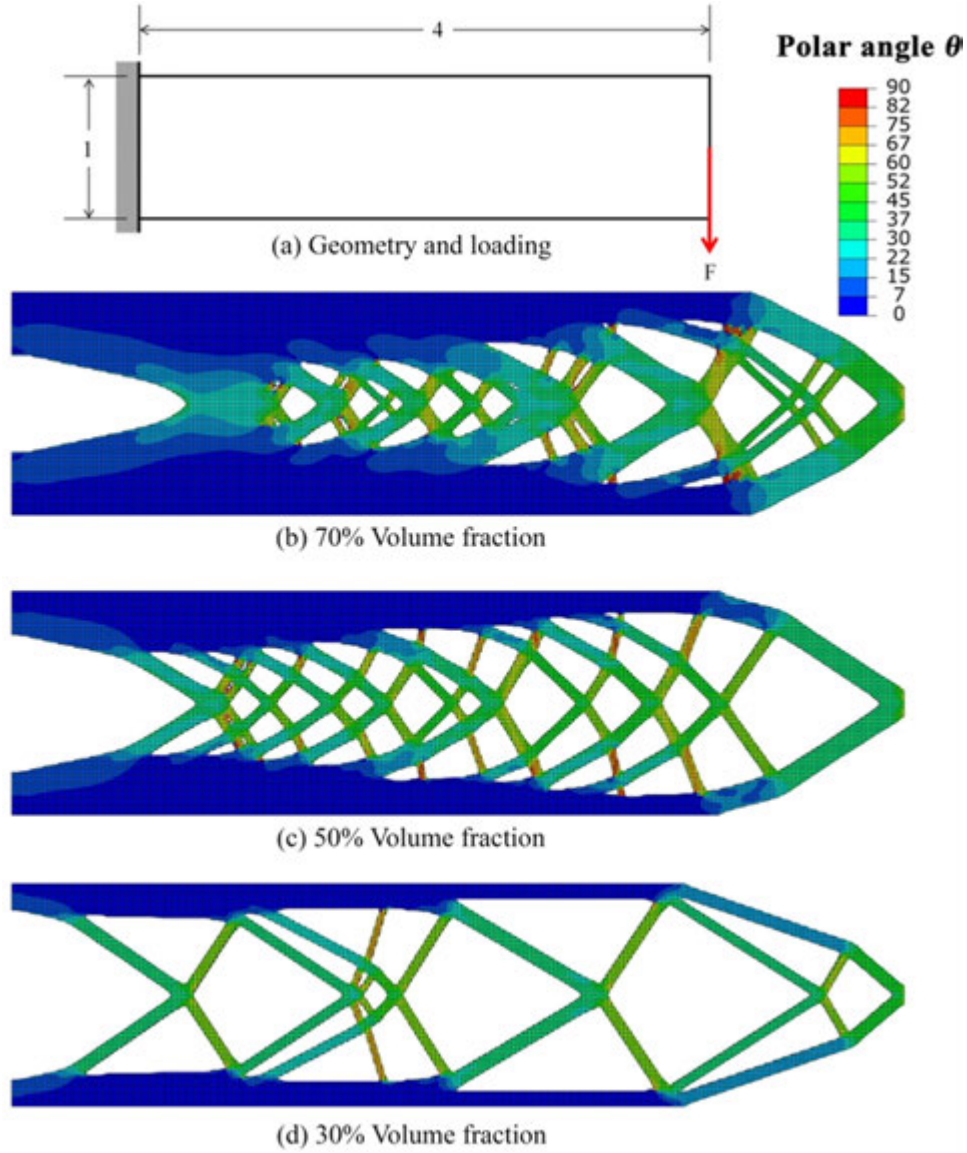


Fig. 3 Optimal topology of symmetric cantilever plate.

Simulation results for test problems and material types are given in Table 4. Fig. 6 shows distributions of density and orientation angle for test problems discussed earlier for UID material; the proposed method is used to determine position of the axis of anisotropy in each element, the initial position of the axis is coincident with OX axis.

Particularly interesting are the final compliance values for UID material, obtained by optimization of density / orientation, compared with initial and final compliance values for materials where density distribution is optimized separately (see Table 4). Here, final compliance values for structures with $\rho_{ev}^\infty = 0.3$ (2D beam) and $\rho_{ev}^\infty = 0.1$ (3D cube, 3D cantilever beam) are close to initial compliance values for structures with $\rho_{ev}^0 = 1.0$. Besides, in case of structures with separately optimized density distribution the final compliance value is 3 times higher than that of “fiber steered” structures. Considering that virtual composites studied here are composed of the same source material by changing its lay-up, the conclusion can be made that by optimizing local reinforcement orientation we can obtain a significantly stiffer structures, compared to those with uniform reinforcement lay-up.

Influence of Algorithm Parameters and Modifications

Algorithm parameters used in test problems were selected when solving 2D beam problems. The number of iterations in all calculations is set to 300. We used various filtering radii $R \in \{0.00, 0.60, 0.80, 1.00, 1.20, 1.42, 1.75, 2.00, 2.25, 2.42\}$ to obtain density distribution $\bar{\rho}_i$ for QIB material. Table 5 shows the final values of mean density and compliance for analyzed filtering radii R . The checkerboard-patterned regions can be observed in some areas at $R \in \{0.00, 0.60, 0.08, 1.00\}$. The size of these regions gets

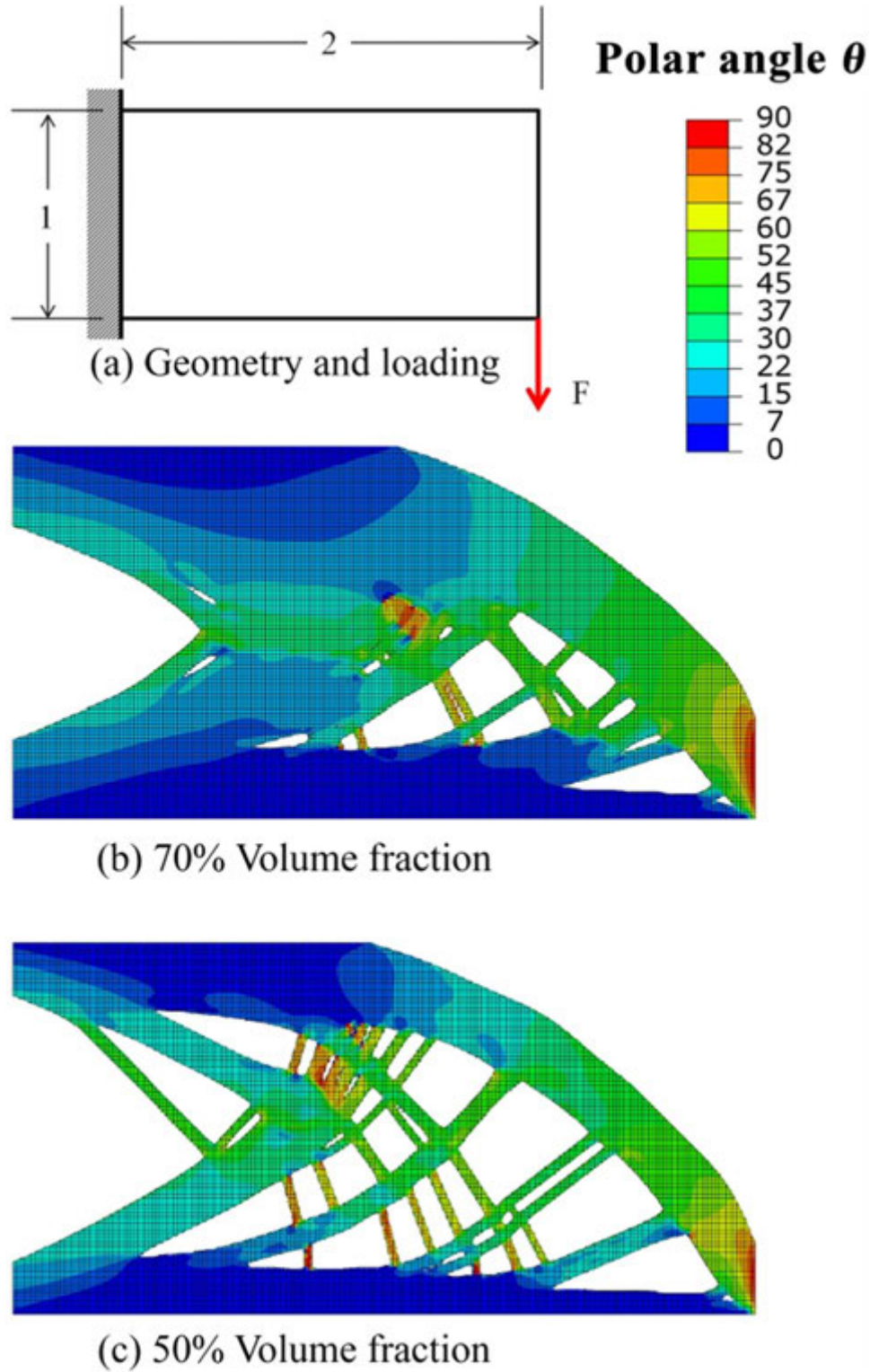


Fig. 4 Optimal topology of symmetric cantilever plate.

smaller as the filtering radius increases. No checkerboard patterns were observed at other values of filtering radius. The increase in filtering radius results in a less detailed structure and, consequently, in increased compliance. To prevent excessive detailization of structures and formation of checkerboard regions the filtering radius of $R = 1.42$ was selected for analysis.

Table 6 shows simulation results for QIB material, obtained with varying parameters k_0 and K that define algorithm convergence rate (11). Various combinations of $k_0 \in \{0.002, 0.010\}$ and $K \in \{1.0, 1.2, 1.4\}$ parameters were tested in the course of the study,

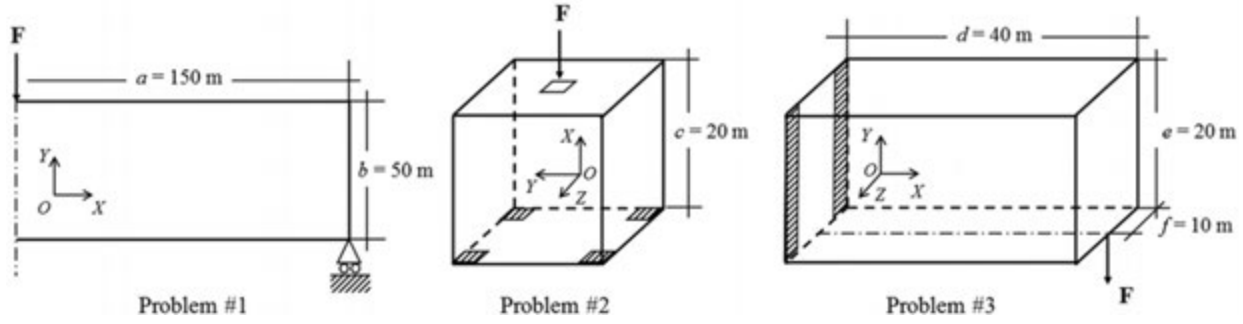


Fig. 5 Test problems: Problem #1 – bending of a 2D beam under concentrated central load (MBB-beam problem); Problem #2 – loading of a 3D cube by vertical load; Problem #3 – loading of a 3D cantilever beam by vertical load.

Table 3 Mechanical properties of materials used in simulation

Properties	Unidirectional reinforced material (UD) (Akkerman, 2002)	Quasi-isotropic laminate (QIL) (Akkerman, 2002)	Quasi-isotropic bulk (QIB)
E_1 (GPa)	100.00	38.99	25.00
E_2 (GPa)	10.00	11.79	25.00
G_{12} (GPa)	3.00	3.21	9.40
ν_{12}	0.30	0.33	0.33
ν_{23}	0.45	0.30	0.33

Table 4 Simulation results for the test problems.

Type of simulation	Initial compliance for initial mean density $\rho_{ev}^0 = 1.0$, J	Final compliance for target mean density ρ_{ev}^∞ , J
2D beam ($\rho_{ev}^\infty = 0.3$)		
QIB material	23.3	60.0
QIL material, plane of isotropy coincides with OXY plane	16.3	44.1
UD material, position of the axis of anisotropy for all finite elements is constant and coincides with OX axis	16.6	50.8
UD material, the proposed method is used to determine position of the axis of anisotropy in each element, initial position of the axis coincides with OX axis	16.6	16.9
3D cube ($\rho_{ev}^\infty = 0.1$)		
QIB material	7.2	31.9
QIL material, plane of isotropy coincides with OXY plane	7.3	40.7
UD material, position of the axis of anisotropy for all finite elements is constant and coincides with OX axis	7.5	54.5
UD material, proposed method is used to determine position of the axis of anisotropy in each element, initial position of the axis coincides with OX axis	7.5	6.5
3D cantilever beam ($\rho_{ev}^\infty = 0.1$)		
QIB material	14.9	27.1
QIL material, plane of isotropy coincides with OXY plane	15.3	26.7
UD material, position of the axis of anisotropy for all finite elements is constant and coincides with OX axis	18.5	31.8
UD material, proposed method is used to determine position of the axis of anisotropy in each element, initial position of the axis coincides with OX axis	18.5	9.8

while other parameters were held constant (see Table 1). To evaluate the rate of convergence in compliance computations we introduce the parameter of $N_{1\%}$, equal to the number of the iteration starting from which the relative difference between current C^n and final C^{300} values of compliance would not exceed 1%, i.e., $\forall n > N_{1\%} : \frac{|C^n - C^{300}|}{C^{300}} 100\% < 1\%$. For $K = 1.0$ the density increment at each iteration step remained constant and equal to $\Delta \rho_i^n = k_0$. In the case of $K = 1.0$ with $k_0 = 0.002$ the algorithm failed to converge to a stable structure within 300 iterations. At $k_0 = 0.01$ the stable structure was obtained within 170 iterations ($N_{1\%} = 170$). The algorithm acceleration technique has allowed a significant reduction in $N_{1\%}$, of up to ($N_{1\%} = 117$), while retaining the final stiffness of the obtained solution $C^{300} = 60.4$ J.

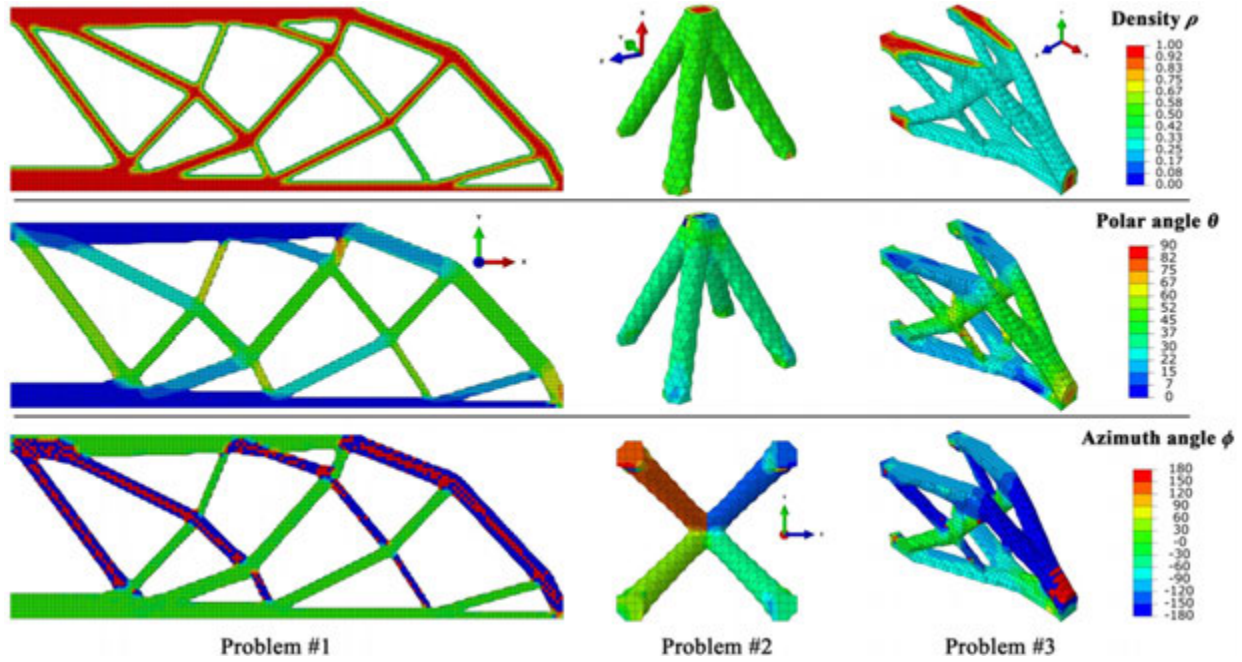


Fig. 6 The final distribution of density $\bar{\rho}$, polar angle θ , and azimuth angle ϕ for the test problems discussed earlier for UD material; the proposed method is used to determine position of the axis of anisotropy in each element, initial position of the axis coincides with OX axis. Where ($0^\circ \leq \theta \leq 90^\circ$) – the polar angle between OX axis and direction of reinforcement, ($-180^\circ \leq \phi \leq 180^\circ$) – the azimuth angle between OY axis and projection of direction of reinforcement onto OYZ plane. Regions with densities exceeding 0.3 are shown, i.e., $\bar{\rho} > 0.3$.

Table 5 Final compliance values for QIB material for different filtering radii R

	Filter radius R									
Compliance C^{300} , J	0.00	0.60	0.80	1.00	1.20	1.42	1.75	2.00	2.25	2.42
	62.5	65.6	68.4	62.5	58.7	60.0	62.8	63.3	65.8	65.5

Table 6 Simulation results for varying k_0 and K parameters determining the algorithm (11) convergence rate for QIB material

k_0	K	C^{300} , J	$N_{I\%}$
0.002	1.0	100.2	None
	1.2	61.0	213
	1.4	62.3	118
0.010	1.0	60.4	170
	1.2	60.4	117
	1.4	60.8	185

Discussion

In this paper we propose the method of finding the optimal distribution of material density and reinforcement orientation within a given design domain. We can use these data as inputs for 3D composite printer instructions that describe the regions with reinforced and non-reinforced plastic, and the orientation of reinforcement by specifying layup direction. At the same time, the study does not cover shell elements. Such elements are common in traditional analysis of composite structures and allow an engineer to define multiple ply layups with interlaced orientations. However, the proposed method can be applied with shell elements, because computations are performed at integration points and the number of points can be specified for each separate ply. In future research we intend to focus at improving the algorithm to account for differences in elastic modulus in tension and compression, local buckling of structural elements, and advanced strength criteria for thermoplastic materials (Fedulov *et al.*, 2017). We also intend to further refine the algorithms with regard to manufacturing constraints imposed by conventional

composite manufacturing processes, such as autoclave forming (Peeters *et al.*, 2015), vacuum infusion (Barnes and Morozov, 2016; Albanesi *et al.*, 2020), winding (Vasiliev *et al.*, 2012), and pultrusion (Safonov *et al.*, 2018; Vedernikov *et al.*, 2020).

Conclusion

A method to optimize the topology of additively manufactured continuous fiber-reinforced composite 3D structures is proposed. The method utilizes a dynamic approach to finding the optimum local distribution of material density. Iterative algorithms for material density optimization have also been developed, with dimensionless algorithm parameters. In order to find the reinforcement vector, the evolutionary algorithm is proposed based on the method of aligning the reinforcement direction with the direction of principal stresses. The algorithms are implemented as a plug-in within ABAQUS environment, via user subroutines Ustdfil, Urdfil, Uexternaldb. To demonstrate the applicability of the method we solve a problem of finding optimal distributions of material density and reinforcement orientation vector for transversely isotropic material with properties typical of continuous fiber-reinforced composites. Following test cases have been studied: bending of a simply supported 2D beam carrying central concentrated load; loading of a 3D cube by vertical load; and bending of a 3D cantilever beam. The proposed algorithm was found to produce lighter structures (by 66% and 90% in case of 2D beam and 3D cube/beam, respectively) with stiffness virtually equivalent to that of initial structures of anisotropic and quasi-isotropic materials. The results obtained can serve as a reference data for future studies. Dimensionless parameters of the algorithm were found, ensuring good convergence of the algorithm and allowing optimization of 3D printed CFR structures to be performed in engineering practice, under service loads.

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Using a Meshless Method to Predict the Strength of Adhesive Single Lap Joints

Luís DC Ramalho and Isidro J Sánchez-Arce, Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal

Raul DSG Campilho and Jorge Belinha, Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal and Polytechnic of Porto, Porto, Portugal

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Nomenclature

b	Body forces	$\mathbf{t}$	External forces applied to Γ_t
c	Constitutive fourth order material tensor	u	Displacement constrains applied to Γ_u
c	Shape parameter	$u(\mathbf{x}_i)$	Displacement of $\mathbf{x}_i$
F	Force vector	$\dot{\mathbf{u}}$	Velocity of a point
Γ	Boundary of Ω	$\mathbf{V}_i$	Influence cell
K	Stiffness matrix	$\mathbf{x}_I$	Interest point
M	Mass matrix	$\mathbf{x}_i$	i^{th} node
$\mathbf{M}_T$	Total moment matrix	$\mathbf{Z}_{ij}$	Null vector
N	Node set		
n	number of nodes		
Ω	Problem domain		
Ω_i	Influence-domain		
$P_i(\mathbf{x}_I)$	Polynomial function of $\mathbf{x}_I$		
p	Shape parameter		
		$\mathbf{t}(\mathbf{x}_I)$	Interpolation function vector
		$\Psi(\mathbf{x}_I)$	By-product vector
		$R_i(\mathbf{x}_I)$	RBF of $\mathbf{x}_I$
		r_{Ii}	Euclidean norm between $\mathbf{x}_I$ and any $\mathbf{x}_i \in \mathbf{V}_I$
		ρ	Material's density
		σ	Stress tensor

Introduction

Modern vehicles have increasingly stricter regulations regarding their emissions. One way to decrease the emissions is to make the vehicles lighter, this can be achieved by using lighter materials in their construction, such as composites, in this department adhesive bonding is also advantageous because it is generally lighter than using bolts or rivets. Additionally, most composites are impossible to join using welding, and bolting or riveting introduce holes into the composite substrates which can lead to delamination, tearing or burrs, (Xu *et al.*, 2018) resulting in weaker components and to premature failures.

With the need for adhesive joints also comes the need to develop accurate analysis techniques which aid in the development of such joints. Before the widespread use of modern computers, closed form analytical solutions were the first type of tools used in the analysis of adhesive joints, some of the most popular of these early methods are the Volkersen model (Volkersen, 1938) and the Hart-Smith model (Hart-Smith, 1973). New types of analytical models continue to be developed, such as the model proposed by Liu *et al.* (2014) in 2014, but nowadays most authors use numerical methods to analyse adhesive joints because they are more versatile and easier to use. The most commonly used numerical method in adhesive joints is the Finite Element Method (FEM), as it happens with most solid mechanics problems. When performing the strength prediction of adhesive joints the failure criteria used are usually divided into three different categories: continuum mechanics, fracture mechanics and damage mechanics criteria.

Continuum mechanics criteria are generally the simplest and easiest to use, but they are also frequently less accurate or mesh dependent since there are stress singularities at the interface corners in most adhesive joints, these criteria assume that the materials are continuous and have no defects or damage. Some recently proposed continuum mechanics criteria were able to predict the strength of adhesive joint with a good degree of accuracy, one such case is the Critical Longitudinal Strain (CLS) criterion, proposed by Ayatollahi and Akhavan-Safar (2015). Fracture mechanics criteria are able to evaluate the stress singularities that occur due to material discontinuities using concepts such as the Stress Intensity Factor (SIF) or its energy equivalent the Strain Energy Release Rate (SERR). Normally these criteria require an initial crack to determine joint strength, that is the case of the work of Goh *et al.* (2013) however some authors propose alternatives that can predict crack initiation, such as the Finite Fracture Mechanics proposed by Leguillon (2002) and used by Hell *et al.* (2014) in adhesive joints. Damage mechanics criteria assume that materials can degrade as their loading increases, leading to a gradual lowering of their stiffness and eventually failure, when they lose all stiffness, Kim and Hong (2018) used damage mechanics to determine joint strength.

In addition to the criteria described previously, Cohesive Zone Models (CZM) can also be used, in fact the review of Ramalho *et al.* (2020a) shows that CZM are currently the most frequently used tool to predict joint strength, an example of their use can be seen in reference (Yousef Kanani *et al.*, 2020). CZM are generally used in combination with the FEM, where CZM are used to simulate the adhesive and continuum elements are used to simulate the substrates. Some authors, such as Machado *et al.* (2019) have also used the eXtended FEM (XFEM) to prediction the strength of adhesive joints, this method is similar to the FEM but enriches the displacement near crack tips.

Meshless methods serve as an alternative to the FEM, one advantage of these methods is that the domain discretization can be performed in a very flexible way, since these methods do not require elements unlike the FEM. There are several meshless methods available, the first one to be developed was the Smoothed Particle Hydrodynamics (SPH), (Lucy, 1977; Gingold and Monaghan, 1977) other popular meshless methods developed since then include the Element-Free Galerkin (EFG) method (Belytschko *et al.*, 1994) and the Reproducing Kernel Particle Method (RKPM) (Chen *et al.*, 1996). However, those methods do not possess the Kronecker delta property, which makes it difficult to impose the boundary conditions. The meshless method used in this work, the Radial Point Interpolation Method (RPIM), (Wang and Liu, 2002) possess the Kronecker delta property, this method was introduced as an improvement of the Point Interpolation Method (PIM) (Liu and Gu, 2001) and the point assembly method (Liu, 2002). There are a few works that used meshless methods in adhesive joints, but their use is very limited when compared with the FEM. Tsai *et al.* (2014) predicted the strength of Double-Cantilever Beams (DCB) using the Symmetric Smoothed Particle Hydrodynamics (SSPH) combined with CZM, achieving accurate strength prediction under mode I and some mixed-mode loadings, but inaccurate strength predictions when mode II loadings were dominant. The RPIM was used by Bodjona and Lessard (2015) to predict the strength of bonded-bolted Single Lap Joints (SLJ), achieving strength predictions similar to the experimental strength, but having some oscillations in the stress fields. The Smoothed Particle Hydrodynamics (SPH) incorporated into Abaqus® was used by Mubashar and Ashcroft (2017) to predict the strength of SLJ, with results comparable to CZM, but the stress along the adhesive mid-thickness line had large oscillations. The Natural Neighbor RPIM (NNRPIM) (Ramalho *et al.*, 2019) and the RPIM (Ramalho *et al.*, 2020b) were used by Ramalho *et al.* along with the CLS criterion to predict the strength of aluminum SLJ, those predictions presented an error lower than 20% for three different adhesives, which is satisfactory considering that it is a continuum mechanics criterion.

The present work aims at using the RPIM to simulate composite SLJ with several overlap lengths (L_O), bonded with three different adhesives, to compare the stresses and strains obtained with this method with the ones obtained using the FEM. Additionally, in this work the CLS criterion was used to predict the strength of the joints and those predictions were compared to the experimental strength of the joints, to check if the choice of L_O used to determine the critical parameters has an impact on the strength predictions, and if so, determining if there is a general rule that can be used which will provide the most accurate strength predictions.

Formulation

Radial Point Interpolation Method

As in the FEM, a geometry has to be discretised in order to analyse it using the RPIM. In this case, it is discretised by a nodal cloud, instead of elements. The geometry represents the domain (Ω), which contains a set of nodes $\mathbf{N} = \{x_1, x_2, \dots, x_n\} \in \mathbb{R}^3$, where n is the number of nodes inside Ω .

Subsequently, a background grid has to be created; whether or not this background grid coincides with the nodal distribution is not important. The grid itself only aids the numerical integration of the governing equations (Liu, 2003; Belinha, 2014). On the other hand, as these methods are based on the nodal distributions, the nodal interaction is determined by the concept of influence-domains (Ω_i), which looks radially for the closer nodes to every interest point. It has been suggested that influence domains with 9–16 nodes are appropriate for the RPIM (Belinha *et al.*, 2016). An example of how these influence domains are determined and how they overlap with each other is shown in Fig. 1. Then, the relationships between the nodes are determined by interpolation functions as it is described below.

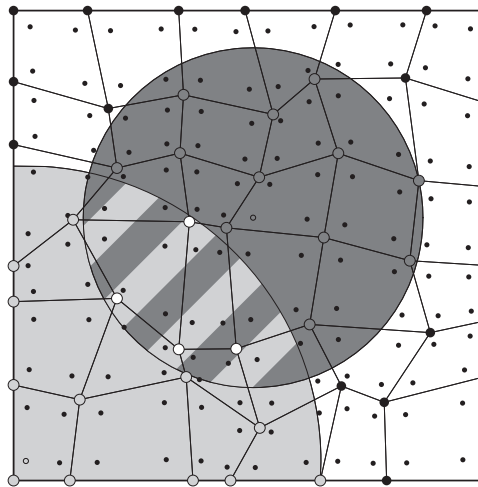


Fig. 1 Example of two overlapping influence domains.

Then, the relationships between the nodes are determined by interpolation functions. Considering a point of interest $\mathbf{x}_1$, which is surrounded by neighboring nodes $\mathbf{x}_i \in \Omega_1$. Then, the value of a field variable, like the displacement u , at $\mathbf{x}_1$ (i.e., $u(\mathbf{x}_1)$), can be determined by the sum of two functions, one radial-based ($R_i(\mathbf{x}_1)$) and one polynomial ($P_j(\mathbf{x}_1)$), (Belinha et al., 2016) as shown in Eq. (1):

$$u(\mathbf{x}_1) = \sum_{i=1}^n R_i(\mathbf{x}_1) a_i(\mathbf{x}_1) + \sum_{j=1}^m P_j(\mathbf{x}_1) b_j(\mathbf{x}_1), \quad (1)$$

where $a_i(\mathbf{x}_1)$ and $b_j(\mathbf{x}_1)$ are the non-constant coefficients of the radial basis function (RBF) and the polynomial function (P), respectively. From Eq. (1), it can be seen that both non-linear coefficients are vectors with respective lengths n and m , which correspond to the number of nodes in the domain and to the number of polynomial terms employed. Belinha et al. (2016) suggested that $m = 1$ is appropriate. The RBF used is the multi-quadratics function, which is shown in Eqs. (2a) and (2b):

$$R(r_{li}) = (r_{li}^2 + c^2)^p \quad (2a)$$

$$r_{li} = \sqrt{(x_i - x_1)^2 + (y_i - y_1)^2 + (z_i - z_1)^2} \quad (2b)$$

where r_{li} is the norm between $\mathbf{x}_1$ and the nodes belonging to the Ω_1 . Values for the shape parameters c and p were optimized previously, taking the values of $c = 1.42$ and $p = 1.03$, as suggested in References (Wang and Liu, 2002; Belinha et al., 2016). Subsequently, by applying the interpolation function Eq. (1) to $\mathbf{N}$ the values of $a_i(\mathbf{x}_1)$ and $b_j(\mathbf{x}_1)$ are determined (Belinha et al., 2016; Farahani et al., 2019). Hence, Eq. (1) could be represented matricially, as follows:

$$\mathbf{u}_s = \mathbf{R}\mathbf{a} + \mathbf{P}\mathbf{b}, \quad (3)$$

where

$$\mathbf{R} = \begin{bmatrix} r_1(\mathbf{x}_1) & r_2(\mathbf{x}_1) & \cdots & r_n(\mathbf{x}_1) \\ r_1(\mathbf{x}_2) & r_2(\mathbf{x}_2) & \cdots & r_n(\mathbf{x}_2) \\ \vdots & \vdots & \ddots & \vdots \\ r_1(\mathbf{x}_n) & r_2(\mathbf{x}_n) & \cdots & r_n(\mathbf{x}_n) \end{bmatrix} \quad (4)$$

$$\mathbf{P} = \begin{bmatrix} p_1(\mathbf{x}_1) & p_2(\mathbf{x}_1) & \cdots & p_m(\mathbf{x}_1) \\ p_1(\mathbf{x}_2) & p_2(\mathbf{x}_2) & \cdots & p_m(\mathbf{x}_2) \\ \vdots & \vdots & \ddots & \vdots \\ p_1(\mathbf{x}_n) & p_2(\mathbf{x}_n) & \cdots & p_m(\mathbf{x}_n) \end{bmatrix} \quad (5)$$

Then, Eqs. (1) and (3) can be rearranged as follows:

$$u(\mathbf{x}_1) = \left\{ \mathbf{r}(\mathbf{x}_1)^T; \mathbf{p}(\mathbf{x}_1)^T \right\} \mathbf{M}_T^{-1} \left\{ \begin{matrix} \mathbf{u}_s \\ \mathbf{z} \end{matrix} \right\} = \left\{ \varphi(\mathbf{x}_1)^T; \Psi(\mathbf{x}_1)^T \right\} \left\{ \begin{matrix} \mathbf{u}_s \\ \mathbf{z} \end{matrix} \right\} \quad (6)$$

where $\mathbf{M}_T$ is the moment matrix:

$$\mathbf{M}_T = \begin{bmatrix} \mathbf{R} & \mathbf{P} \\ \mathbf{P}^T & \mathbf{Z} \end{bmatrix} \quad (7a)$$

$$Z_{ij} = 0 \quad (7b)$$

Thus, Eq. (7a) could be used to obtain a solution using matricial techniques, as described by (Belinha, 2014). Hence, Eq. (6) can be expressed in terms of the shape function value ($\varphi_i(\mathbf{x}_1)$) of $\mathbf{x}_1$, (Belinha, 2014) as follows:

$$u(\mathbf{x}_1) = \sum_{i=1}^n \varphi_i(\mathbf{x}_1) u_i, \quad (8)$$

Afterwards, material properties, boundary conditions ($\mathbf{u}$), and loads ($\bar{\mathbf{t}}$) are introduced. Material properties correspond to the material's constitutive tensor ($\mathbf{c}$), which relates stress and strain in the elastic regime. Then, a Lagrangian is calculated by considering an equilibrium of the energies (based on the Galerkin weak form), as described by Eq. 9 (Belinha, 2014).

$$L = \frac{1}{2} \int_{\Omega} \rho \dot{\mathbf{u}}^T \dot{\mathbf{u}} d\Omega - \frac{1}{2} \int_{\Omega} \boldsymbol{\varepsilon}^T \boldsymbol{\sigma} d\Omega + \int_{\Omega} \mathbf{u}^T \mathbf{b} d\Omega + \int_{\Gamma_t} \mathbf{u}^T \mathbf{t} d\Gamma, \quad (9)$$

where $\dot{\mathbf{u}}$, Ω , and ρ represent the velocity, volume (the domain here), and the material's density, respectively. Then, Eq. (9) has to be minimized:

$$L = \int_{\Omega} \boldsymbol{\sigma} d\boldsymbol{\varepsilon} d\Omega - \int_{\Omega} \mathbf{b} \cdot \mathbf{u} d\Omega - \int_{\Gamma_t} \bar{\mathbf{t}} \cdot \mathbf{u} d\Gamma + \rho \int_{\Omega} (\mathbf{u}^T \dot{\mathbf{u}}) d\Omega = 0, \quad (10)$$

It can be seen that Eq. (10) is a function of $\mathbf{u}$, even the first term. Thus, by means of Eq. (8) it could be related to nodal displacement. Moreover, the derivative of Eq. (10) would provide the stiffness matrix ($\mathbf{K}$), the force vector ($\mathbf{F}$), and the mass matrix ($\mathbf{M}$), (Belinha, 2014) as shown in Eq. (11):

$$\mathbf{K}\mathbf{u} + \mathbf{M}\ddot{\mathbf{u}} = \mathbf{F}, \quad (11)$$

Finally, the boundary conditions (natural and essential) can be applied in as similar way as in the FEM. Once the equation systems were assembled and the boundary conditions and loads imposed, the solution procedure is similar to the one of the FEM, as it has been described by Liu (2003); Belinha *et al.* (2016); Liu and Gu (2005).

Critical Longitudinal Strain Criterion

The CLS criterion was first proposed by Ayatollahi and Akhavan-Safar (2015) inspired by the theory of critical distances of Taylor (2008). This criterion has only been applied to SLJ, the joints analysed in this work, and it determines that they fail when the longitudinal strain (ε_{xx}) reaches a critical value (ε_c) at a critical distance (r_c) in the adhesive mid-thickness line. The determination of the critical parameters is made using experiments and numerical simulations, both are necessary. Usually, these experiments and simulations need to be performed to SLJ with two distinct overlap lengths (L_O), but Akhavan-Safar *et al.* (2017) proposed a formula that allows the determination of the critical parameters by testing a single L_O , however, this formula is only applicable to some adhesives, so it was not used in this work.

The procedure to determine the critical parameters, ε_c and r_c , is as follows: first, it is necessary to perform experiments on SLJ with two different L_O and their failure loads are registered; then, linear-elastic numerical simulations are performed to the joints tested previously, with the experimentally determined failure loads imposed as boundary conditions; afterwards, the ε_{xx} along the adhesive mid-thickness line is extracted from both simulations and they are plotted superimposed, with the distance normalized over the L_O ; finally, the intersection point between those two curves is determined, which allows the determination of the critical parameters, the ε_{xx} of this intersection point is the ε_c and its x/L_O is the r_c . Having determined the critical parameters, it is then possible to predict the strength of similar SLJ with different L_O by performing numerical simulations and determining the force necessary to reach the ε_c at r_c .

Results

Geometry and Materials

The SLJ simulated were all made out of a unidirectional carbon-epoxy pre-preg (SEAL® Texipreg HS 160 RM) with 0.15 mm thickness plies and a $[0]_{16}$ lay-up. The elastic orthotropic properties of the ply are presented in Table 1, obtained from Campilho *et al.* (2013).

The composite substrates were bonded with three different types of adhesives: the brittle epoxy Araldite® AV138, the moderately ductile epoxy Araldite® 2015 and the ductile polyurethane Sikaforce® 7888. The mechanical properties of these adhesives were obtained from Neto *et al.* (2012) and Campilho *et al.* (2013) being presented in Table 2. The SLJ geometry is presented in Fig. 2, all joints possessed the same geometrical dimensions with the exception of the L_O which varied from 10 mm to 80 mm in increments of 10 mm, which means that 8 different joints were tested with each adhesive, in the figure the gray zones represent the adhesive and the white zones represent the substrates.

Table 1 Ply mechanical properties

$E_{xx} = 1.09 \times 10^5$ MPa	$\nu_{xy} = 0.342$	$G_{xy} = 4315$ MPa
$E_{yy} = 8819$ MPa	$\nu_{xz} = 0.342$	$G_{xz} = 4315$ MPa
$E_{zz} = 8819$ MPa	$\nu_{yz} = 0.380$	$G_{yz} = 3200$ MPa

Table 2 Adhesive mechanical properties

Properties	AV138	2015	7888
E (GPa)	4.89 ± 0.81	1.85 ± 0.21	1.89 ± 0.81
ν	0.35^a	0.33^a	0.33^a
σ_y (MPa)	36.49 ± 2.47	12.63 ± 0.61	13.20 ± 4.83
σ_f (MPa)	39.45 ± 3.18	21.63 ± 1.61	28.60 ± 2.0
ε_f	1.21 ± 0.10	4.77 ± 0.15	43.0 ± 0.6
G (GPa)	1.56 ± 0.01	0.56 ± 0.21	0.71^b
τ_y (MPa)	25.1 ± 0.33	14.6 ± 1.3	—
τ_f (MPa)	30.2 ± 0.40	17.9 ± 1.8	20^a
γ_f (MPa)	7.8 ± 0.7	43.9 ± 3.4	100^a
G_{IC} (N/mm)	0.20^b	0.43 ± 0.02	1.18 ± 0.22
G_{IIC} (N/mm)	0.38^b	4.70 ± 0.34	8.72 ± 1.22

^aManufacturer data

^bEstimated by Campilho *et al.* (2011).

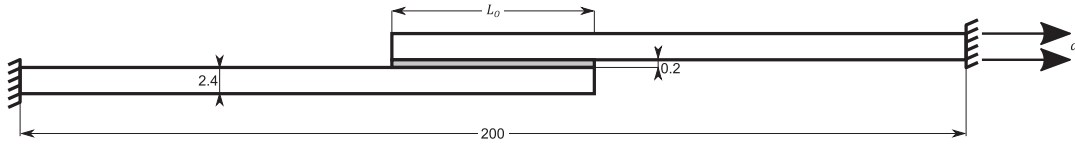


Fig. 2 Geometry and boundary conditions of the SLJ.

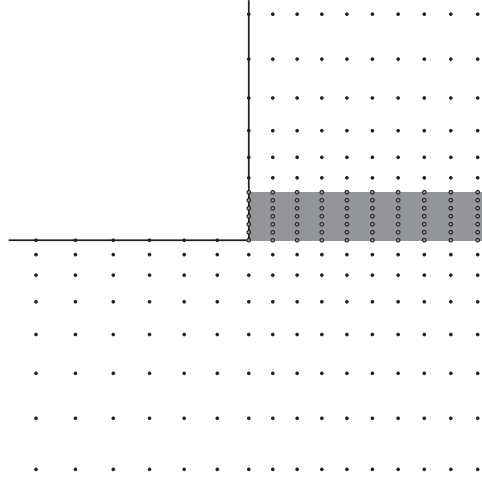


Fig. 3 Nodal density and biases near lower interface corner of the $L_0 = 30$ mm joint.

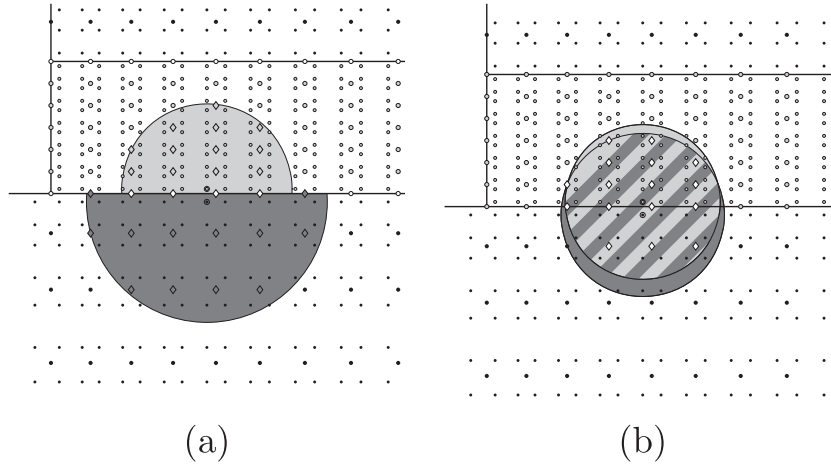


Fig. 4 Influence domains in the interface region using the restriction (a) and without using the restriction (b).

Discretization

The different L_0 were discretized following some general rules, namely, every discretization has 5 lines of nodes along the adhesive thickness and 12 lines along the substrate thickness, in addition to the 2 interface lines, there are also biases along the overlap length and the substrate thickness to ensure that there is a higher nodal density near the interface corners which are the most critical zones in SLJ. These features and their intended purpose can be observed in Fig. 3, where the discretization near one of the interfacial corners is shown for the joint with $L_0 = 30$ mm, the discretizations for the other L_0 are similar, but they have more nodes as L_0 increases. Every simulation was made assuming small deformations, linear-elastic material behavior and plane-strain.

Since this geometry has a material interface, the RPIM influence domains in the regions near the interfaces need a special rule. This rule consists in creating a restriction which disallows the influence domains on one of the materials to penetrate the

other material, as it can be seen in Fig. 4(a) where the influence domain of the encircled adhesive node is shown in light gray, and the influence domain of the encircled substrate node is shown in dark gray, the nodes belonging to both domains are represented by white diamonds. A solution similar to this was also adopted by Liu (2010) for the Element Free Galerkin (EFG) method to deal with this problem. If these influence domains behave like any other influence domain it leads to influence domains of one material penetrating the other material. In extreme cases it can even lead to those influence domains being composed of more nodes of the other material than of nodes of the integration point's material, as is seen in Fig. 4(b), this problem is much more present when the discretization of one material is much finer than the discretization of the other material, as is the case here.

Stress and Strain Distribution

Firstly, it is important to verify the validity of using the influence domain restriction near the interfaces, as opposed to letting the influence domains be created normally in that region. To that end, the peel stress (σ_{yy}) and the shear stress (τ_{xy}) at the interface of both options are compared to each other and to a solution obtained with the FEM, using the same number of nodes, since the FEM is a well established method. The results of this comparison are shown in Fig. 5, where it can be seen that adopting the influence domain restriction has a significant influence on both stress components and the results using the restriction are closer to the FEM results than the results not using the influence domain restriction. Since this comparison shows that the influence domain restriction yields better results, it was used in all further simulations.

The σ_{yy} , τ_{xy} and ϵ_{xx} along the adhesive mid-thickness plane obtained using the RPIM are also compared with the FEM results for all L_O tested. This comparison is only shown for the Araldite® AV138 in Fig. 6 for the sake of brevity, but it is similar for the other adhesives, it shows that the differences between the two numerical methods are very small. The FEM simulations were performed using quadrilateral elements with 4 nodes and the same total number of nodes. These curves also show that the σ_{yy} and the τ_{xy} peak very close to the overlap ends and the magnitude of these peaks increases with the L_O . The τ_{xy} also shows a decrease in the middle of the overlap as L_O increases, there is also a compressive stress in the middle of the overlap for small L_O , which moves closer and closer to the overlap ends as the L_O increases, even if there is no compressive stress in the middle of the overlap, there is always a lack of a peeling stress.

Strength Prediction

When using the CLS criterion to determine joint strength it is first necessary to determine the critical parameters using the process described in Section "Critical Longitudinal Strain Criterion". In this work this determination was performed using every possible L_O combination, which means that the critical parameters were determined with 28 different L_O combinations, for every adhesive. This will be useful to verify if the chosen L_O combination has an influence on the critical parameters and if the conclusions of Ramalho *et al.* (2020b) regarding the ideal choice of L_O combination to determine the critical parameters remains valid when the SLJ have a different geometry and different substrates, composite substrates instead of aluminum. An example of how the critical parameters for one of the L_O combination using Araldite® AV138 is shown in Fig. 7(a), where the

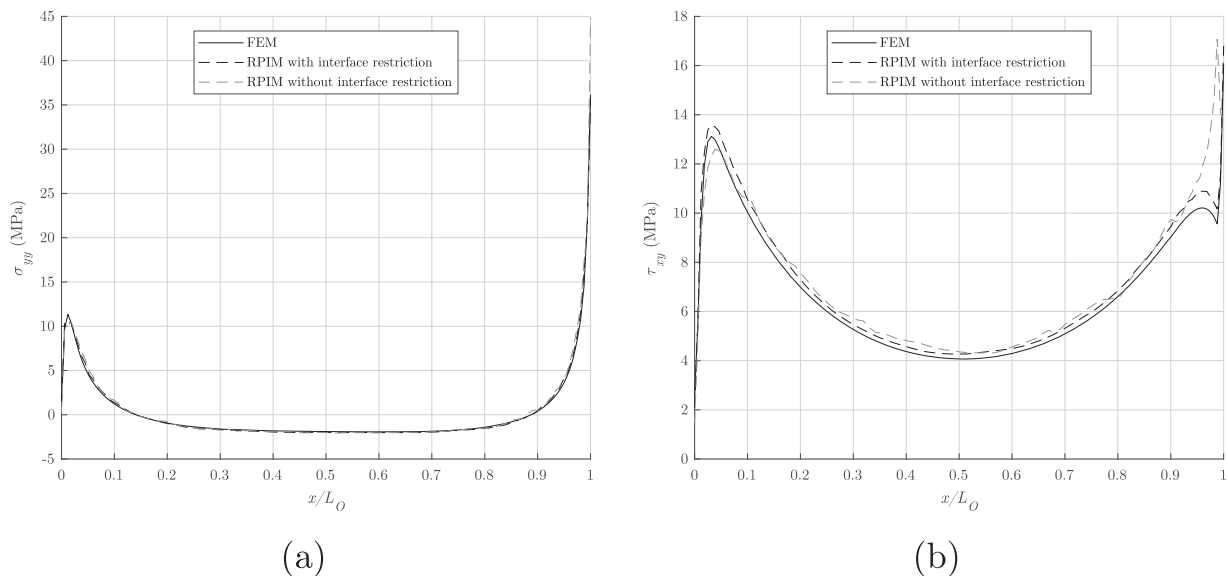


Fig. 5 σ_{yy} (a) and τ_{xy} (b) comparison at the lower interface for joints bonded with Araldite® AV138 and $L_O = 10$ mm.

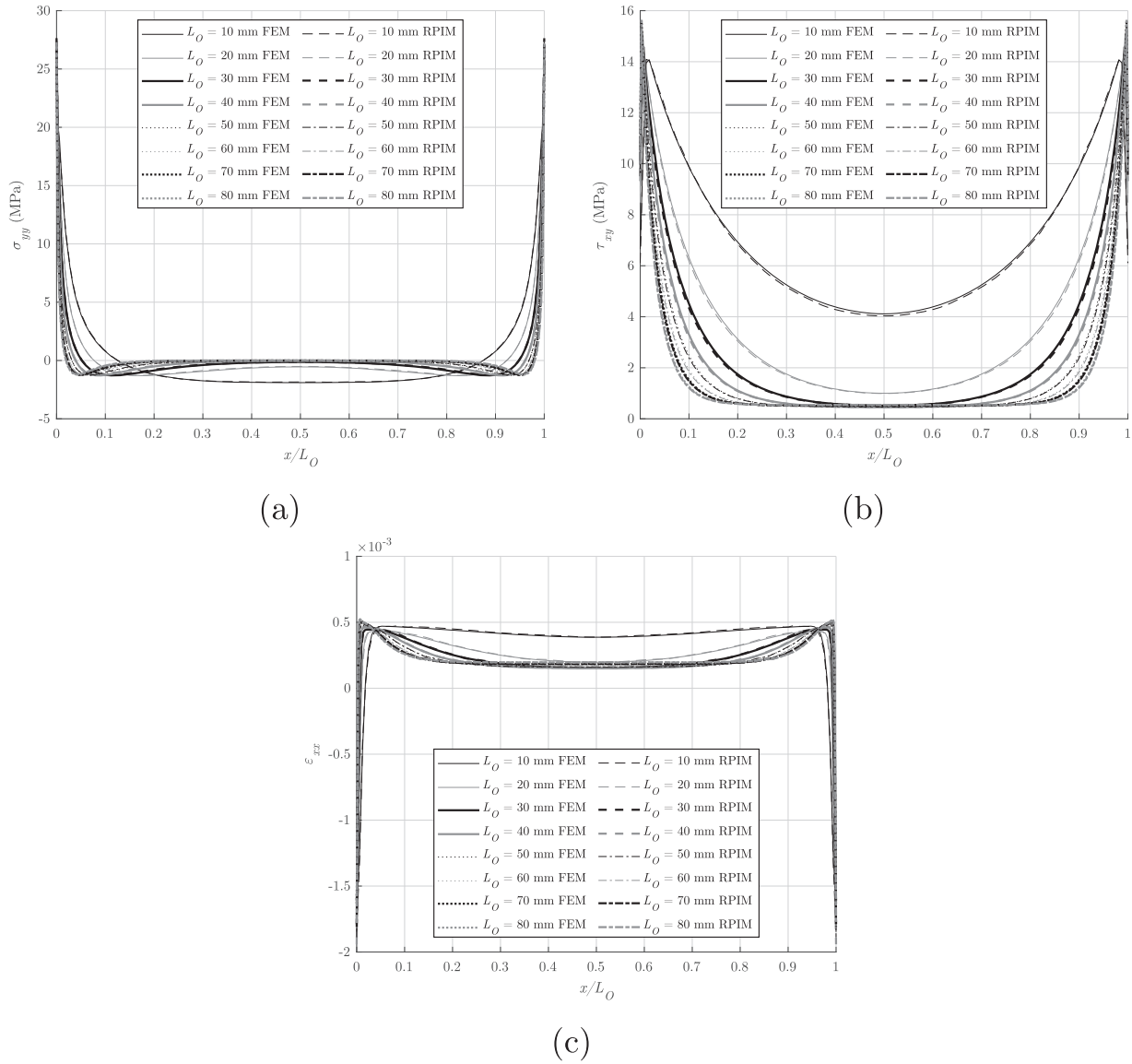


Fig. 6 σ_{yy} (a), τ_{xy} (b) and ε_{xx} (c) along the adhesive mid-thickness plane for joints bonded with Araldite® AV138.

ε_{xx} and x/L_O of the black point, representing the intersection between the two curves, are ε_c and r_c , respectively. In later steps the strength prediction for other L_O using those critical parameters was performed by extracting the force required to reach ε_c at r_c , as is shown in Fig. 7(b).

Tables 3, 4 and 5 show the critical parameters for the three different adhesives, for every L_O combination with which it was possible to determine the critical parameters. From those tables it can be seen that with some L_O combinations the critical parameters were not determined, this is because their ε_{xx} curves did not intersect, an example of this is shown in Fig. 7(c), a problem that was not observed when using aluminum substrates in reference Ramalho *et al.* (2020b). For the brittle Araldite® AV138 only with one L_O combination it was not possible to determine the critical parameters, $L_{O1} = 70$ mm and $L_{O2} = 80$ mm, which means that for the other 27 combinations the critical parameter were determined without any issue. The same cannot be said about the more ductile adhesives, for the Araldite® 2015 it was only possible to determine the critical parameters with two different L_O combinations and for the Araldite® 7888 this was possible with nine different L_O combinations, this means that for these two adhesives it was impossible to determine the critical parameters with most L_O combinations.

Tables 3, 4 and 5 also present the average strength prediction error when using the critical parameters obtained with each L_O combination. From that data, it can be concluded that the rule of Ramalho *et al.* (2020b) of using the smallest L_O as L_{O1} and the largest L_O as L_{O2} to obtain the best strength prediction is not true when the SLJ are made of composite substrates instead of aluminum substrates. The strength predictions for the Araldite® AV138 present a generally low average error, regardless of the L_O

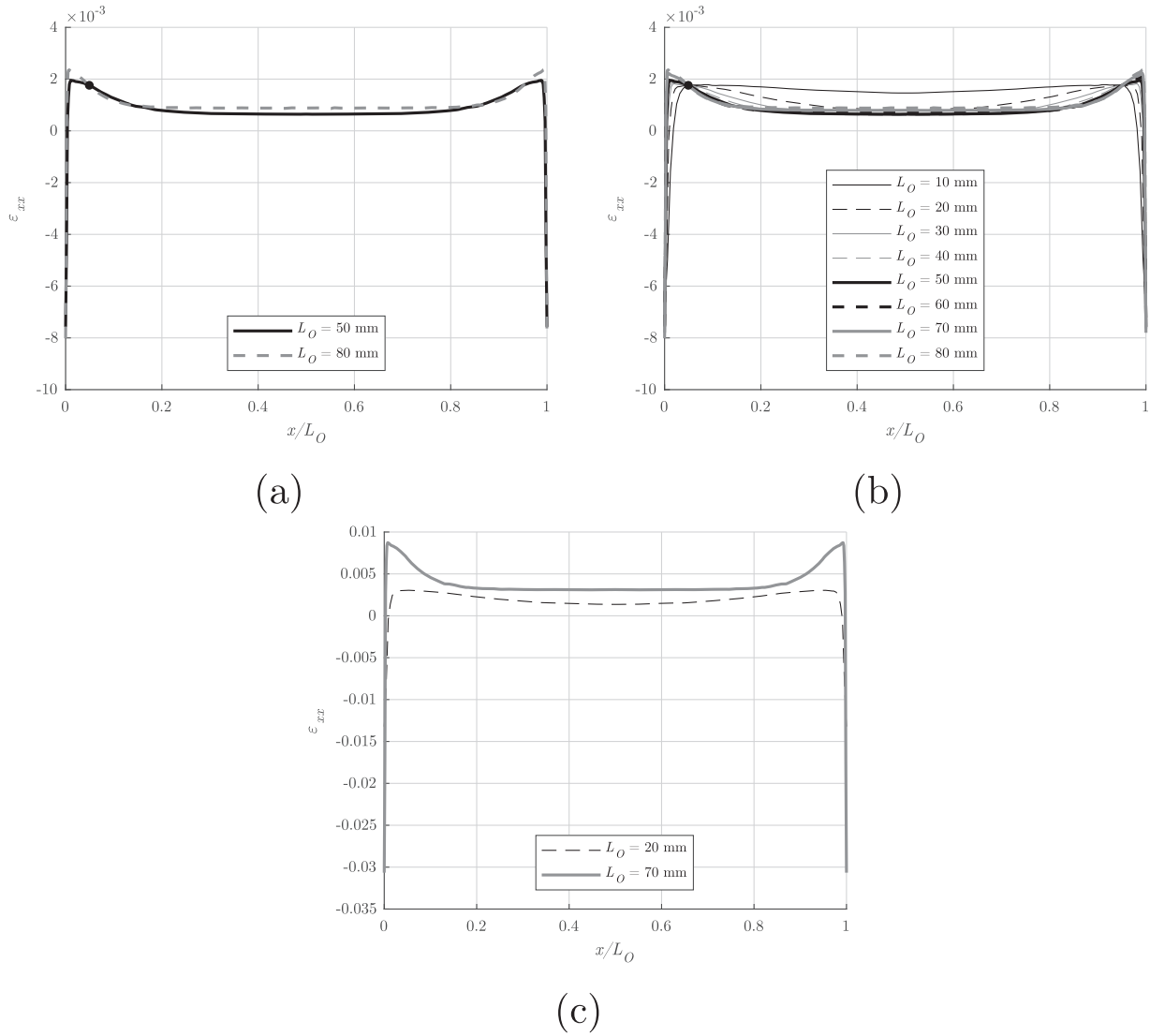


Fig. 7 Determining the intersection between the ε_{xx} curves of one L_O combination for the Araldite® AV138 (a), ε_{xx} of all L_O when those critical parameters are reached (b), one of the L_O combinations for the Araldite® 2015 where no intersection occurred (c).

combination used to determine the critical parameters. In most cases the average strength prediction error is lower than 10%, or very close to it, except for when $L_{O1} = 10$ mm and $L_{O2} = 20$ mm, and $L_{O1} = 60$ mm and $L_{O2} = 70$ mm, where the errors are 15.77% and 20.33%, respectively. Overall, the best strength predictions were obtained using $L_{O1} = 50$ mm and $L_{O2} = 80$ mm to determine the critical parameters. For the Araldite® 2015, as was discussed previously, it was only possible to obtain the critical parameters with 2 of the 28 different L_O combinations, which means that it was only possible to perform the strength predictions using those 2 different critical parameters. The average errors of the strength predictions using those critical parameters are similar, but when using $L_{O1} = 10$ mm and $L_{O2} = 30$ mm the average error is slightly smaller. For the Sikaforce® 7888, the strength predictions always present an average error higher than 20%, regardless of the L_O combination used to determine the critical parameters. For this adhesive the best strength predictions were achieved when using $L_{O1} = 50$ mm and $L_{O2} = 60$ mm to determine the critical parameters.

The strength predictions obtained with the critical parameters that yielded the lowest average error for each adhesive are shown in Fig. 8 and compared to the experimental strength. There it can be seen that for the Araldite® AV138, the strength predictions are always within the experimental variance, except for $L_O = 20$ mm, but in that case the difference is small. For the Araldite® 2015, the strength predictions are close to the experimental values when $L_O \leq 40$ mm, but for larger L_O the CLS criterion predicts a strength plateau when in reality the strength increased almost linearly with the L_O . For the Sikaforce® 7888, the strength predictions are close to the experimental strength when $L_O \geq 30$ mm, but for the two smallest L_O the joint strength is greatly over-predicted, being almost double the experimental strength when $L_O = 10$ mm.

Table 3 Critical parameters and average strength prediction errors using them for the Araldite® AV138

L_{01} (mm)	L_{02} (mm)	ε_c	r_c	Average strength prediction error
10	20	0.034	1.79×10^{-3}	15.77%
	30	0.037	1.67×10^{-3}	10.14%
	40	0.043	1.79×10^{-3}	4.68%
	50	0.044	1.80×10^{-3}	4.66%
	60	0.047	1.84×10^{-3}	6.35%
	70	0.043	1.79×10^{-3}	4.65%
	80	0.045	1.83×10^{-3}	5.40%
20	30	0.076	1.58×10^{-3}	10.74%
	40	0.076	1.58×10^{-3}	10.77%
	50	0.064	1.60×10^{-3}	5.76%
	60	0.063	1.61×10^{-3}	5.50%
	70	0.055	1.61×10^{-3}	6.22%
	80	0.057	1.61×10^{-3}	5.48%
30	40	0.077	1.58×10^{-3}	10.81%
	50	0.062	1.63×10^{-3}	5.44%
	60	0.061	1.63×10^{-3}	5.10%
	70	0.052	1.66×10^{-3}	5.35%
	80	0.055	1.65×10^{-3}	4.66%
40	50	0.047	1.78×10^{-3}	4.51%
	60	0.053	1.74×10^{-3}	5.07%
	70	0.043	1.79×10^{-3}	4.65%
	80	0.049	1.77×10^{-3}	4.41%
50	60	0.057	1.69×10^{-3}	4.95%
	70	0.041	1.82×10^{-3}	6.13%
	80	0.049	1.76×10^{-3}	4.32%
60	70	0.197	0.81×10^{-3}	20.33%
	80	0.040	1.91×10^{-3}	9.91%

Table 4 Critical parameters and average strength prediction errors using them for the Araldite® 2015

L_{01} (mm)	L_{02} (mm)	ε_c	r_c	Average strength prediction error
10	20	0.279	1.08×10^{-3}	25.82%
	30	0.292	1.07×10^{-3}	25.92%

Table 5 Critical parameters and average strength prediction errors using them for the Sikaforce® 7888

L_{01} (mm)	L_{02} (mm)	ε_c	r_c	Average strength prediction error
10	20	0.269	1.88×10^{-3}	28.37%
	30	0.329	1.81×10^{-3}	28.25%
40	50	0.123	3.91×10^{-3}	23.91%
	60	0.124	3.90×10^{-3}	23.74%
	80	0.082	4.90×10^{-3}	38.67%
50	60	0.126	3.84×10^{-3}	22.97%
	80	0.058	6.03×10^{-3}	57.26%
60	70	0.074	5.56×10^{-3}	50.14%
	80	0.041	7.12×10^{-3}	82.16%

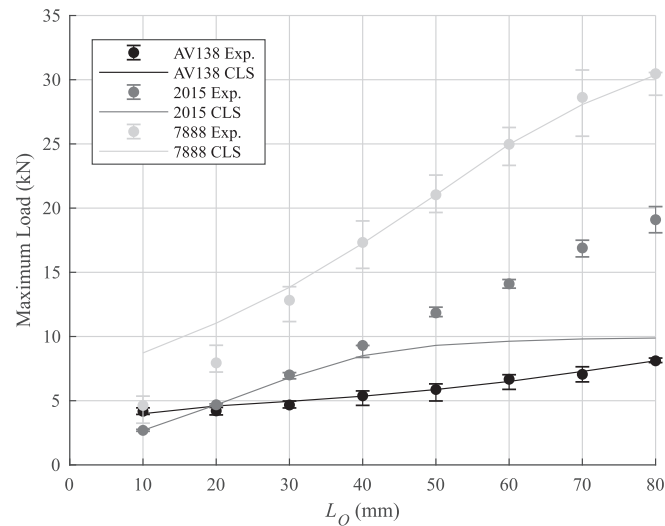


Fig. 8 Strength predictions for all L_O using the critical parameters with the smallest average error.

Conclusion

One of the goals of this work was to simulate adhesively bonded SLJ made of composite substrates using the RPIM, to assess if this meshless method can be applied to this type of problem. The composite substrates have anisotropic mechanical properties, which adds complexity to the simulation, especially considering that this is coupled with an isotropic material, the adhesives. However, the RPIM shown no real difficulties when dealing with that, with the exception of the influence domains near the material interfaces, which was easily dealt with by restricting the influence domains in that region. When comparing to the FEM, the stress components along the adhesive mid-thickness line obtained with the RPIM were shown to be similar, validating this method for this type of problem.

Additionally, this work also wanted to assess the use of the CLS criterion when performing the strength prediction of SLJ made with composite substrates simulated with the RPIM, and to determine if there is a general rule that can be followed when determining the critical parameters. It was found that there is no discernible rule for the determination of the critical parameters that consistently yield more accurate strength predictions across the three adhesives. For the brittle Araldite® AV138, the strength predictions were generally accurate, regardless of the L_O combination used to determine the critical parameters, while for the ductile adhesives the strength prediction yielded average errors higher than 20%, regardless of the L_O combination used to determine the critical parameters, and for most L_O combinations it was not possible to determine the critical parameters because there was no intersection between the ε_{xx} curves. These findings contrast with the findings of Ramalho *et al.* (2020b) for SLJ with the same adhesives and aluminum substrates, instead of composite substrates, where it was found that using the smallest L_O as L_{O1} and the largest L_O as L_{O2} for all adhesives resulted the best strength predictions. Overall, the CLS criterion can give accurate strength prediction, however its use is limited because the strength predictions can have great variations depending on the L_O combination used to determine the critical parameters, except for the most brittle adhesive where the strength predictions are good for almost all L_O combinations.

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Homogenizing the Elastic Properties of Composite Material Using the NNRPIM

Daniel ES Rodrigues, Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal

Jorge Belinha, Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal and Polytechnic of Porto, Porto, Portugal

Francisco MA Pires, Renato MN Jorge, and Lúcia MJS Dinis, Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal and Faculty of Engineering of the University of Porto, Porto, Portugal

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Introduction

Meshless methods discretize the problem domain in a set of nodes, independent from each other. Therefore, unlike the FEM, the concept of mesh, as their designation reveals, is inexistent. Thus, to ensure the nodal connectivity, there is an overlap rule of influence-domains where the field variables are interpolated. In meshless formulations, influence-domains can be areas or volumes centered on an interest point, containing a fixed or variable number of nodes from the nodal mesh. Meshless methods' interpolation functions have virtually a higher order, allowing a higher continuity and reproducibility than the FEM. Due to being mesh independent, the refinement procedure is easier and re-meshing issues – which usually appear in the FEM when the problem deals with transitory geometry – are not verified in meshless methods. Regarding the numerical integration, it can be performed using nodal dependent background integration meshes.

The first meshless method was proposed in 1977: the Smooth Particle Hydrodynamics Method (SPH) (Gingold and Monaghan, 1977). In 1994, the first global weak form-based meshless method was presented, the Element Free Galerkin Method (EFGM) (Belytschko *et al.*, 1994). Since then, several other meshless methods were proposed, based on strong and weak forms, using different shape functions, distinct integration schemes, etc. Some relevant meshless methods are the Meshless Local Petrov-Galerkin Method (MLPG) (Atluri and Zhu, 1998), the Reproducing Kernel Particle Method (RKPM) (Liu *et al.*, 1995), Point Interpolation Method (PIM) (Liu and Gu, 2001), the Point Assembly Method (Liu, 2001), the Radial Point Interpolation Method (RPIM) (Wang and Liu, 2002) or the Natural Neighbor Radial Point Interpolation Method (NNRPIM) (Belinha, 2014; Dinis *et al.*, 2007). The NNRPIM was born from the combination of the Radial Point Interpolators (RPI) with the natural neighbors' geometric concept. Like the RPIM, the Point Assembly Method and the PIM, the NNRPIM is an interpolant meshless method. This means the shape functions possess the delta Kronecker property, making the imposition of the natural and essential boundary conditions easier.

Although the NNRPIM is a recently developed meshless method (it was firstly proposed in 2007 (Dinis *et al.*, 2007)), it was extended to many fields in computational mechanics such as the static analysis of isotropic and composite plates and beams (Dinis *et al.*, 2008; Dinis *et al.*, 2010b; Dinis *et al.*, 2011; Belinha *et al.*, 2013a,b; Moreira *et al.*, 2014), the 3D shell-like approach for laminated plates and shells (Dinis *et al.*, 2010a), anisotropic elasto-plastic analysis (Moreira *et al.*, 2016), nonlinearity problems (Dinis *et al.*, 2009), fracture mechanics (Azevedo *et al.*, 2015; Belinha *et al.*, 2016; Belinha *et al.*, 2017), biomechanical problems (Belinha, 2014; Belinha *et al.*, 2012), dynamic applications (Dinis *et al.*, 2009b), large deformations (Dinis *et al.*, 2009a), etc. In this work, the NNRPIM is combined with a material homogenization technique in order to obtain homogenized elastic properties of composite materials.

The NNRPIM – Natural Neighbor Radial Point Interpolation Method

The Natural Neighbor Radial Point Interpolation Method (NNRPIM) relies on geometrical and mathematical constructions to obtain the nodal and integration meshes, which are dependent on each other. Therefore, the NNRPIM can be considered a truly meshless method since there is a dependency between the integration mesh and the spatial relations between the nodes. The NNRPIM makes use of radial point interpolation functions, which are a combination of multiquadric radial basis functions (Hardy, 1990a) – firstly proposed by Hardy (Hardy, 1990b) – and polynomial basis functions. The RPI shape functions possess compact support and the delta Kronecker property. To establish the discrete system of equations, the NNRPIM uses the Galerkin weak form.

In the following sections, some geometrical concepts are introduced to describe the NNRPIM procedure. Then, the NNRPIM interpolation functions are obtained, and the meshless discrete system of equations is established.

Natural Neighbors' Concept, Nodal Connectivity, and Numerical Integration

A generic meshless method procedure starts with the discretization of the problem domain in a set of nodes. The nodal mesh can be regular or irregular, with the last one having, in general, lower accuracy. Then, using the Voronoï diagram, influence-cells are created. In Fig. 1, it is described the construction of the Voronoï diagram from an unstructured set of nodes. Considering a set of N distinct nodes (Fig. 1(a)), the Voronoï diagram of this set of nodes is composed by sub-regions, closed and convex, called Voronoï cells (whose constructions are schematized in Fig. 1(b)). Each Voronoï cell represents the geometric region whose points

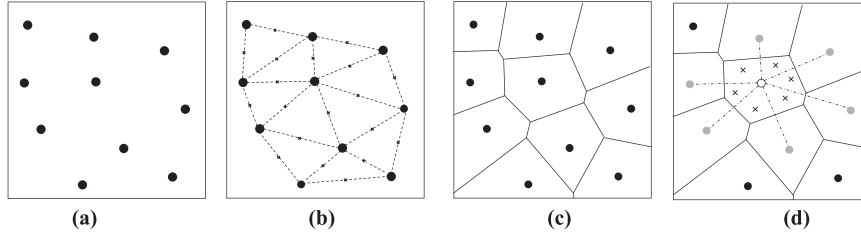


Fig. 1 (a) set of nodes. (b) Construction of Voronoï cells. (c) Full Voronoï diagram. (d) First-degree influence-cell of the white node containing the nodes represented in gray; Construction of the integration mesh.

are closer to the central node than any other node outside of that cell. This way, the assemblage of the complete set of Voronoï cells defines the Voronoï diagram (Fig. 1(c)). The Voronoï cells do not overlap each other neither leave any gaps between them.

The Voronoï diagram can be used to establish influence-cells (Belinha, 2014), based on the natural neighbor's concept. Considering an interest node x_l , the influence-cell of x_l can be established by the gathering of the nodes whose Voronoï cell is adjacent to the Voronoï cell of x_l – as exemplified in Fig. 1(d). This is the definition of natural neighbors: the nodes whose Voronoï cells possess common edges are called natural neighbors. The influence-cell described in Fig. 1(d) is called a first-degree influence-cell since it contains just the first natural neighbors of a node. Nevertheless, second-degree influence-cells can also be established considering the nodes belonging to the first-degree influence-cell as well as the first natural neighbors of all those nodes within the first-degree influence-cell. Generally, this latter configuration of influence-cell allows to obtain more accurate solutions but the computation cost is significantly higher (Belinha, 2014).

The establishment of a Voronoï diagram for a set of nodes also permits the construction of a background integration mesh. As shown in Fig. 1(d), using the Delaunay triangulation, the area of a Voronoï cell is divided into several sub-areas called sub-cells. Then, using the Gauss-Legendre numerical scheme, it is possible to place an integration point on the geometric center of each sub-cell and use their area as integration weight. The procedure described is called an Order 0 integration scheme (Belinha, 2014) which, according to the literature, is sufficient to perform the numerical integration of the discrete system of equations.

Interpolation Functions

In the NNRPIM, the shape functions used to interpolate the field variables within each influence-cell are a combination of multiquadric radial basis functions (MQ-RBF) (Wang and Liu, 2002; Hardy, 1990a) and polynomial basis functions. Thus, considering an interest point x_l , the displacement field in x_l can be obtained using: $u(x_l) = \sum_{i=1}^n \varphi_i(x_l) u_i$, where n the number of nodes within the influence-cell of x_l , u_i is the value of the field variable in the node i and $\varphi_i(x_l)$ represents the value of the interpolation function associated with the node i calculated at the interest point x_l (Belinha, 2014). Knowing the nature of the interpolation functions, the equation of interpolation can be written in the following form (Dinis et al., 2007; Belinha, 2014; Belinha et al., 2010):

$$u(x_l) = R^T(x_l) a(x_l) + p^T(x_l) b(x_l) \quad (1)$$

In Eq. (1), $R(x_l) = \{R_1(x_l), R_2(x_l), \dots, R_n(x_l)\}^T$ is the vector whose components are the multiquadric RBFs given by: $R_i(x_l) = R(r_{li}) = [r_{li}^2 + c^2]^p$, where r_{li} is the Euclidian distance between the interest point x_l and the neighbor node x_i (obtained using: $r_{li} = |x_i - x_l| = \sqrt{(x_l - x_i)^2 + (y_l - y_i)^2}$), and c and p shape parameters. The vector $p(x_l) = \{p_1(x_l), p_2(x_l), \dots, p_m(x_l)\}^T$, with m components, contains the polynomial basis calculated in x_l , being m the basis monomial number. Finally, vectors $a(x_l) = \{a_1(x_l), a_2(x_l), \dots, a_n(x_l)\}^T$ and $b(x_l) = \{b_1(x_l), b_2(x_l), \dots, b_m(x_l)\}^T$ contain the non-constant coefficients of $R_i(x_l)$ and $p_j(x_l)$, respectively.

If a polynomial basis function is used in the establishment of the interpolation functions, another condition needs to be verified to ensure a unique approximation (Belinha, 2014): $\sum_{i=1}^n p_j(x_i) a_i(x_i) = 0 \Leftrightarrow p^T(x_i) a(x_i) = 0$, $j = \{1, 2, \dots, m\}$, which combined with Eq. (1) results in the system (2):

$$\begin{Bmatrix} u_s \\ 0 \end{Bmatrix} = \begin{bmatrix} R & p \\ p^T & 0 \end{bmatrix} \begin{Bmatrix} a \\ b \end{Bmatrix} = G \begin{Bmatrix} a \\ b \end{Bmatrix} \quad (2)$$

being u_s the vector containing the nodal parameters of the field variable, u , for each node within the support domain of the RPI shape function (i.e., within the influence-cell of an interest point x_l): $u_s = \{u_1, u_2, \dots, u_n\}^T$. The radial moment matrix R , with dimensions $[n \times n]$, is defined as $R_{ij} = R(r_{ij})$:

$$R = \begin{bmatrix} R(r_{11}) & R(r_{12}) & \dots & R(r_{1n}) \\ R(r_{21}) & R(r_{22}) & \dots & R(r_{2n}) \\ \vdots & \vdots & \ddots & \vdots \\ R(r_{n1}) & R(r_{n2}) & \dots & R(r_{nn}) \end{bmatrix} \quad (3)$$

And the polynomial moment matrix, $\mathbf{p}$, with dimensions $[n \times m]$:

$$\mathbf{p} = \begin{bmatrix} p_1(\mathbf{x}_1) & p_2(\mathbf{x}_1) & \dots & p_m(\mathbf{x}_1) \\ p_1(\mathbf{x}_2) & p_2(\mathbf{x}_2) & \dots & p_m(\mathbf{x}_2) \\ \vdots & \vdots & \ddots & \vdots \\ p_1(\mathbf{x}_n) & p_2(\mathbf{x}_n) & \dots & p_m(\mathbf{x}_n) \end{bmatrix} \quad (4)$$

being each line defined as $\mathbf{p}_i = \{1, x, y, x^2, xy, y^2, \dots\}$, $i = \{1, 2, \dots, n\}$, and $\{\mathbf{x}_1, \mathbf{x}_2, \dots, \mathbf{x}_n\}$ a set of interest points generating n different influence-cells. Thus, to obtain the non-constant coefficients, $\mathbf{a}(\mathbf{x}_I)$ and $\mathbf{b}(\mathbf{x}_I)$, Eq. (2) can be rewritten:

$$\begin{Bmatrix} \mathbf{a}(\mathbf{x}_I) \\ \mathbf{b}(\mathbf{x}_I) \end{Bmatrix} = \mathbf{G}^{-1} \begin{Bmatrix} \mathbf{u}_s \\ \mathbf{0} \end{Bmatrix} \quad (5)$$

and substituting Eq.(5) in Eq. (1),

$$\mathbf{u}(\mathbf{x}_I) = \{\mathbf{R}^T(\mathbf{x}_I), \mathbf{p}^T(\mathbf{x}_I)\} \mathbf{G}^{-1} \begin{Bmatrix} \mathbf{u}_s \\ \mathbf{0} \end{Bmatrix} = \{\Phi^T(\mathbf{x}_I), \Psi^T(\mathbf{x}_I)\} \begin{Bmatrix} \mathbf{u}_s \\ \mathbf{0} \end{Bmatrix} \quad (6)$$

where vector $\Phi^T(\mathbf{x}_I) = \{\varphi_1(\mathbf{x}_I), \varphi_2(\mathbf{x}_I), \dots, \varphi_n(\mathbf{x}_I)\}$ contains the interpolation functions calculated at the interest point $\mathbf{x}_I$. The vector $\Psi^T(\mathbf{x}_I) = \{\Psi_1(\mathbf{x}_I), \Psi_2(\mathbf{x}_I), \dots, \Psi_m(\mathbf{x}_I)\}$ is a by-product vector without relevant meaning.

Meshless Discrete System of Equations for Static Solid Mechanics Problems

Consider a solid domain Ω with boundary Γ , where $\Gamma \in \Omega : \Gamma_u \cup \Gamma_t = \Gamma$, being Γ_u the essential boundary and Γ_t the natural boundary. The natural boundary conditions are defined as $\boldsymbol{\sigma} \mathbf{n} = \bar{\mathbf{t}}$ (being $\boldsymbol{\sigma}$ the Cauchy stress tensor, $\mathbf{n}$ the unit normal vector to the boundary, and $\bar{\mathbf{t}}$ the traction on the natural boundary), and the essential boundary conditions are defined as $u = \bar{u}$ (being $\mathbf{u}$ a kinematically admissible displacement field, and $\bar{u}$ the displacement imposed). For a linear-static problem, the equilibrium equations can be expressed as follows:

$$\nabla \boldsymbol{\sigma} + \mathbf{b} = \mathbf{0} \quad (7)$$

where ∇ is the gradient operator and $\mathbf{b}$ are the body forces. In NNRPIM, to establish the meshless discrete system of equations, it is used the Galerkin weak form:

$$\int_{\Omega} \boldsymbol{\sigma} \cdot \delta \boldsymbol{\varepsilon} d\Omega - \int_{\Omega} \delta \mathbf{u}^T \cdot \mathbf{b} d\Omega - \int_{\Gamma_t} \delta \mathbf{u}^T \cdot \bar{\mathbf{t}} d\Gamma_t = 0 \quad (8)$$

where $\delta \boldsymbol{\varepsilon}$ is the virtual work deformation and $\delta \mathbf{u}$ is the virtual displacement. The virtual strain tensor $\delta \boldsymbol{\varepsilon}$ can be obtained using $\delta \boldsymbol{\varepsilon} = \mathbf{B} \cdot \delta \mathbf{u}$, where $\mathbf{B}$ is the deformation matrix, which establishes the relationship between the displacements and the strain. Matrix $\mathbf{B}$ can be expressed as:

$$\mathbf{B}(\mathbf{x}_I) = \mathbf{L} \cdot \mathbf{H}(\mathbf{x}_I) = \begin{bmatrix} \frac{\partial}{\partial x} & 0 \\ 0 & \frac{\partial}{\partial y} \\ \frac{\partial}{\partial y} & \frac{\partial}{\partial x} \end{bmatrix} \cdot \begin{bmatrix} \varphi_1(\mathbf{x}_I) & 0 \\ 0 & \varphi_1(\mathbf{x}_I) \\ \varphi_2(\mathbf{x}_I) & 0 \\ 0 & \varphi_2(\mathbf{x}_I) \\ \dots & \dots \\ \varphi_n(\mathbf{x}_I) & 0 \\ 0 & \varphi_n(\mathbf{x}_I) \end{bmatrix}^T \quad (9)$$

where $\mathbf{L}$ is a differential operator written for a generic two-dimensional problem, and matrix $\mathbf{H}$ represents blocks of diagonal matrices, $\mathbf{H}^j$, containing the interpolation function of each node j of the influence-cell of $\mathbf{x}_I$, with $\mathbf{H}^j = \varphi_j(\mathbf{x}_I) \mathbf{I}$, being $\mathbf{I}$ an identity matrix with dimension $[d \times d]$, where d is the number of degrees of freedom of the analyzed problem (in Eq. (9), $d = 2$).

Introducing the stress-strain relationship, $\boldsymbol{\sigma} = \mathbf{c} \boldsymbol{\varepsilon}$, and knowing that $\boldsymbol{\varepsilon} = \mathbf{B} \cdot \mathbf{u}$, Eq. (8) can be rewritten:

$$\delta \mathbf{u}^T \int_{\Omega} \mathbf{B}^T \mathbf{c} \mathbf{B} \mathbf{u} d\Omega = \delta \mathbf{u}^T \int_{\Omega} \mathbf{H}^T \mathbf{b} d\Omega + \delta \mathbf{u}^T \int_{\Gamma_t} \mathbf{H}^T \bar{\mathbf{t}} d\Gamma_t \quad (10)$$

Removing the virtual displacement $\delta \mathbf{u}$ from Eq. (10), it is finally obtained the meshless discrete system of equations:

$$\int_{\Omega} \mathbf{B}^T \mathbf{c} \mathbf{B} d\Omega \mathbf{u} = \int_{\Omega} \mathbf{H} \mathbf{b} d\Omega + \int_{\Gamma_t} \mathbf{H} \bar{\mathbf{t}} d\Gamma_t \quad (11)$$

which is known in its standard form, $\mathbf{K} \mathbf{u} = \mathbf{F}$, with $\mathbf{K} = \int_{\Omega} \mathbf{B}^T \mathbf{c} \mathbf{B} d\Omega$ being the stiffness matrix, and $\mathbf{F} = \int_{\Omega} \mathbf{H} \mathbf{b} d\Omega + \int_{\Gamma_t} \mathbf{H} \bar{\mathbf{t}} d\Gamma_t$ the loading vector.

Thus, in the NNRPIM formulation, after the construction of the interpolation functions regarding each influence-cell, the numerical integration takes place, following the description previously given. Then, the local system of equations is established to be assembled into a global discrete system of equations. The assemblage process is based on the nodal connectivity provided by

the overlap of influence-cells. Then, the Gauss elimination method (Belinha, 2014) is used to obtain the solution of the meshless discrete system of equations.

Homogenization Technique

Fiber composite materials find several applications in the engineering field, particularly in the aircraft and automotive industries. Due to their high specific mechanical properties, these materials are nowadays widely used in primary structural components. Therefore, the prediction of their mechanical behavior is essential to prevent failure due to cracking, delamination, buckling, etc. On the other hand, these materials are anisotropic and heterogeneous which turns challenging the study of their behavior.

Two approaches can be followed when analyzing these materials: the macromechanical analysis and the micromechanical analysis. In the first approach, the material is treated as a homogeneous orthotropic continuum (Devireddy and Biswas, 2014). In the second approach, the micro-heterogeneities are considered since they influence the mechanical properties of the material and, on a larger scale, they have a great influence on its macroscopic behavior (Carvalho, 2016). The macroscale analysis cannot be as accurate as the microscale analysis when the material possesses a complex microstructure.

In micromechanics, it is usual to make use of a Representative Volume Element (RVE), which statistically represents the microstructure of the material. Being L_{macro} the size of the macroscale structure, L_{RVE} the size of the RVE, and L_{het} the size of the RVE's heterogeneities, the following relation leads to the definition of scale separation (Hashin, 1983): $L_{het} \ll L_{RVE} \ll L_{macro}$. The RVE, containing micro-heterogeneities and different material constituents (in composite materials, there are matrix and fibers within a given volume fraction), represents an infinitesimal point of the macroscale. Thus, using the RVE, it is possible to obtain at microscale homogenized material properties which are mechanically equivalent to the heterogeneous material properties of the RVE domain. Consequentially, using a scale transition technique, a mechanical analysis can be simplified by considering that an infinitesimal point of the macroscale (which may contain several micro-heterogeneities, gaps, etc) has the homogenized material properties calculated from the RVE. This homogenization technique allows to have a direct relationship between the micro and the macroscale and results in more reliable constitutive models.

In this work, the NNRPIM formulation, previously presented, is combined with an homogenization technique to extract homogenized elastic properties of composite materials in a plane-strain state. In the following sections, the scale transition theory, the averaging theory, the Hill-Mandel principle, and the imposition of periodic boundary conditions are presented. Gathering all these concepts, in the end, the homogenization procedure is explained.

Scale Transition Theory and Microscopic Equilibrium Problem

Consider the point $\mathbf{x}$ at the macroscale with coordinates $\mathbf{x} = \{x_1 \ x_2 \ x_3\}$ associated with an RVE with volume V – Fig. 2.

At the microscale, there are γ material points that compose the RVE domain. A perturbation of the RVE equilibrium is translated by a local deformation at the macroscale in $\mathbf{x}$. The displacement $\mathbf{u}(\mathbf{x})$ is expressed as:

$$\mathbf{u}(\mathbf{x}) = \mathbf{X} - \mathbf{x} \quad (12)$$

where $\mathbf{X}$ represents the coordinates of the point $\mathbf{x}$ after the perturbation of the RVE and, therefore, after the deformation of the macroscopic point. Assuming a motion φ which transforms $\mathbf{x}$ into $\mathbf{X}$ such that $\varphi(\mathbf{x}) = \mathbf{X}$, it is possible to express the displacement $\mathbf{u}(\mathbf{x})$ as a function of the motion: $\mathbf{u}(\mathbf{x}) = \varphi(\mathbf{x}) - \mathbf{x}$. Under these conditions, a macroscopic deformation gradient can be defined as a

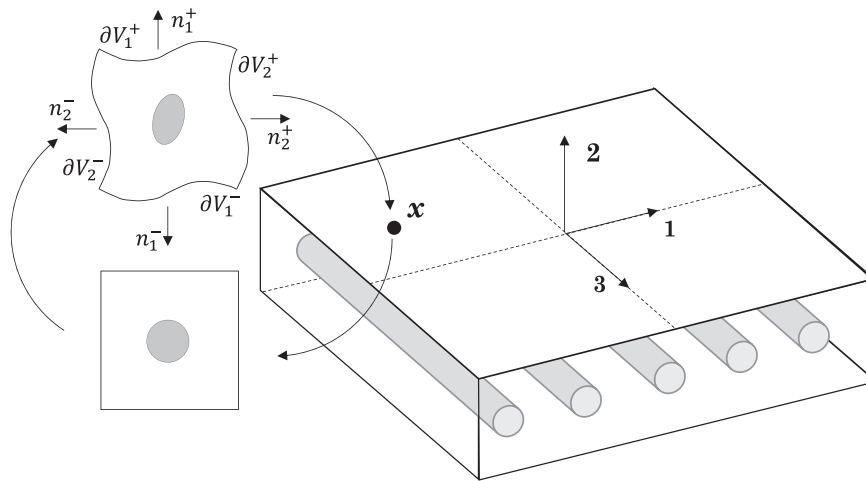


Fig. 2 Schematic representation of the multi-scale procedure: composite laminated plate at the macroscale, RVE at the microscale, imposition of periodic boundary conditions to solve the microscopic equilibrium problem.

second order tensor of the motion φ :

$$F(\mathbf{X}) = \nabla_{\mathbf{x}} \varphi(\mathbf{x}) = \frac{\partial \mathbf{X}}{\partial \mathbf{x}} \quad (13)$$

which can be rewritten in the following way, considering that $\mathbf{X} = \mathbf{x} + \mathbf{u}(\mathbf{x})$,

$$F(\mathbf{X}) = \mathbf{I} + \nabla_{\mathbf{x}} \mathbf{u}(\mathbf{x}) \quad (14)$$

being $\mathbf{I}$ the second-order identity tensor. Introducing now the numerical homogenization procedure:

$$F(\mathbf{X}) = \frac{1}{V} \int_V F(\mathbf{Y}) dV \quad (15)$$

where $F(\mathbf{Y}) = \mathbf{I} + \nabla_{\mathbf{x}} \mathbf{u}(\mathbf{Y})$ is a microscopic deformation gradient, and $\mathbf{Y}$ is the deformed version of the point whose undeformed configuration was previously defined as $\mathbf{y}$. The homogenization procedure consists in calculating the macroscopic computational fields through the volume average of their microscopic corresponding fields over the RVE. Thus,

$$F(\mathbf{X}) = \frac{1}{V} \int_V [\mathbf{I} + \nabla_{\mathbf{x}} \mathbf{u}(\mathbf{Y})] dV = \mathbf{I} + \frac{1}{V} \int_V \nabla_{\mathbf{x}} \mathbf{u}(\mathbf{Y}) dV \quad (16)$$

Applying a macroscopic deformation gradient to the RVE generates a microscopic displacement field $\mathbf{u}(\mathbf{Y})$. This field can be decomposed as follows:

$$\mathbf{u}(\mathbf{Y}) = (F(\mathbf{X}) - \mathbf{I})\mathbf{y} + \tilde{\mathbf{u}}(\mathbf{Y}) \quad (17)$$

where the first parcel depends on the prescribed deformation gradient and the initial configuration of the point $\mathbf{y}$ and the second parcel is the displacement fluctuation field, $\tilde{\mathbf{u}}(\mathbf{Y})$, an unknown variable of the microscopic equilibrium problem. Consequentially, the microscopic deformation gradient can be defined in terms of the macroscopic counterpart and the displacement fluctuation field:

$$\begin{aligned} F(\mathbf{Y}) &= \mathbf{I} + \nabla_{\mathbf{x}} [(F(\mathbf{X}) - \mathbf{I})\mathbf{y} + \tilde{\mathbf{u}}(\mathbf{Y})] \\ &= F(\mathbf{X}) + \nabla_{\mathbf{x}} \tilde{\mathbf{u}}(\mathbf{Y}) \end{aligned} \quad (18)$$

Hill-Mandel Principle

As described by Eq. (17), the imposition of specific macroscale gradient to the RVE allows to obtain a microscopic displacement field, $\mathbf{u}(\mathbf{Y})$. Thus, using this displacement field, it is possible to compute strain and stress microscopic fields for the RVE domain, knowing the strain-displacement and stress-strain relationships previously mentioned. Using the averaging theory – already stated with Eq. (15) – the macroscopic strain, $\bar{\epsilon}$, and stress, $\bar{\sigma}$, fields are obtained:

$$\begin{aligned} \bar{\sigma}_{ij} &= \frac{1}{V} \int_V \sigma_{ij} dV \\ \bar{\epsilon}_{ij} &= \frac{1}{V} \int_V \epsilon_{ij} dV \end{aligned} \quad (19)$$

As stated by the Hill-Mandel Principle using energy considerations, the output of Eq. (19) is that the homogeneous medium of a composite given by the average stresses and strains is equivalent to its actual heterogeneous medium (Sun and Vaidya, 1996). Thus, the Hill-Mandel principle establishes the relationship between the two scales: if the sub-scale-modeling is energetically consistent, the deformation energy at the macroscopic level should be equal to the volume average of micro-scale stress power (Nguyen *et al.*, 2012):

$$\bar{\sigma}_{ij} \delta \bar{\epsilon}_{ij} = \frac{1}{V} \int_V \sigma_{ij} \delta \epsilon_{ij} dV \quad (20)$$

which can be rewritten in terms of the RVE boundary traction force (considering an absence of body forces) knowing that $\delta \epsilon_{ij} = \delta \bar{\epsilon}_{ij} + \delta \tilde{\epsilon}_{ij}$ and $\frac{1}{V} \int_V \sigma_{ij} \delta \tilde{\epsilon}_{ij} dV = 0$, and using the Gauss Theorem,

$$\frac{1}{V} \oint_{\partial V} t_i \delta \tilde{u}_i dS = 0 \quad (21)$$

being t_i an external traction and ∂V represents the boundary of the RVE, as seen in Fig. 2. With the previous equation, a conclusion can be drawn from the Hill-Mandel Principle: the traction forces are reactions associated to the enforced kinematical constraints applied to $\tilde{\mathbf{u}}(\mathbf{Y})$ at the RVE's boundary.

Periodic Boundary Conditions

To satisfy the condition Eq. (22), the boundary conditions of RVE for the displacement and the traction fields should be properly defined. In the literature, there are several types of boundary conditions that satisfy the Hill-Mandel Principle, such as the linear

displacement boundary condition, uniform traction boundary conditions or periodic boundary conditions (PBCs). According to the literature, the PBCs are the most efficient in terms of convergence rate (Nguyen *et al.*, 2012) and therefore are used in this work.

The kinematical constraint in PBCs require that the boundary of the RVE is always divided into positive and negative parts, as represented in Fig. 2. Each RVE point Y^+ at ∂V^+ has its counterpart point Y^- at ∂V^- . The PBCs can be expressed as follows:

$$\begin{aligned}\tilde{\mathbf{u}}(Y^+) &= \tilde{\mathbf{u}}(Y^-) \\ \tilde{\mathbf{t}}(Y^+) &= -\tilde{\mathbf{t}}(Y^-)\end{aligned}\quad (22)$$

meaning that the displacement fluctuation fields on opposite boundaries of the RVE must be periodically equal and, at the same time, the traction field must be anti-periodic on those opposite boundaries of the RVE. These conditions can be easily imposed in Eq. (11) if a rearrangement of the displacement fluctuation field is made:

$$\tilde{\mathbf{u}} = \begin{Bmatrix} \tilde{\mathbf{u}}^i \\ \tilde{\mathbf{u}}^+ \\ \tilde{\mathbf{u}}^- \end{Bmatrix} \quad (23)$$

where $\tilde{\mathbf{u}}^i$ is the inner nodes' displacement fluctuation, and $\tilde{\mathbf{u}}^+$ and $\tilde{\mathbf{u}}^-$ are the displacement fluctuations on the positive and negative boundaries of the RVE, respectively. Thus, the discrete system of equations can be written in the following standard form for this micromechanical boundary problem:

$$\begin{bmatrix} \mathbf{k}^{ii} & \mathbf{k}^{i+} & \mathbf{k}^{i-} \\ \mathbf{k}^{+i} & \mathbf{k}^{++} & \mathbf{k}^{+-} \\ \mathbf{k}^{-i} & \mathbf{k}^{-+} & \mathbf{k}^{--} \end{bmatrix} \begin{Bmatrix} \tilde{\mathbf{u}}^i \\ \tilde{\mathbf{u}}^+ \\ \tilde{\mathbf{u}}^- \end{Bmatrix} = \begin{Bmatrix} \mathbf{f}^i \\ \mathbf{f}^+ \\ \mathbf{f}^- \end{Bmatrix} \quad (24)$$

According to the space of admissible displacement fluctuations, the first condition of Eq. (23) is enforced using the Gauss elimination method, while the second condition is imposed on the vector of the second member of Eq. (24). Furthermore, to prevent rigid body motions, the nodes at the corners of the RVE were constrained.

Extraction of Homogenized Elastic Properties

As previously mentioned, with the RVE it is possible to obtain effective homogeneous elastic properties which represent the same mechanical behavior as the heterogeneous medium represented in the RVE. In this work, RVEs are used to extract effective elastic properties of a composite material, whose constituents are fibers and a matrix.

Two-dimensional RVEs were considered in this work in a plane strain state since the length of the fibers (whose axial direction is the out-of-plane direction – direction 3 in Fig. 2) is way larger than the in-plane dimensions. Under those conditions, $\varepsilon_{33} = \varepsilon_{32} = \varepsilon_{31} = 0$, but $\sigma_{33} \neq 0$. The Cauchy stress tensor can be expressed as (in the referential $O123$):

$$\boldsymbol{\sigma} = \begin{bmatrix} \sigma_{11} & \sigma_{12} & 0 \\ \sigma_{21} & \sigma_{22} & 0 \\ 0 & 0 & \sigma_{33} \end{bmatrix} \quad (25)$$

The constitutive equation (Reddy, 2004), using the stress and strain tensors in Voigt notation, is:

$$\begin{bmatrix} \sigma_{11} \\ \sigma_{22} \\ \sigma_{33} \\ \tau_{12} \end{bmatrix} = \begin{bmatrix} C_{11} & C_{12} & C_{13} & 0 \\ C_{21} & C_{22} & C_{23} & 0 \\ C_{31} & C_{32} & C_{33} & 0 \\ 0 & 0 & 0 & C_{44} \end{bmatrix} \cdot \begin{bmatrix} \varepsilon_{11} \\ \varepsilon_{22} \\ 0 \\ \gamma_{12} \end{bmatrix} \quad (26)$$

The material constants in Eq. (26) depend on the elastic properties of the material:

$$\begin{aligned} C_{11} &= \frac{1 - \nu_{23}\nu_{32}}{E_2 E_3 \Delta}, & C_{12} &= \frac{\nu_{21} + \nu_{23}\nu_{31}}{E_2 E_3 \Delta}, & C_{13} &= \frac{\nu_{31} + \nu_{21}\nu_{32}}{E_2 E_3 \Delta} \\ C_{21} &= \frac{\nu_{12} + \nu_{32}\nu_{13}}{E_1 E_3 \Delta}, & C_{22} &= \frac{1 - \nu_{13}\nu_{31}}{E_1 E_3 \Delta}, & C_{23} &= \frac{\nu_{32} + \nu_{12}\nu_{31}}{E_1 E_3 \Delta} \\ C_{31} &= \frac{\nu_{31} + \nu_{12}\nu_{23}}{E_1 E_2 \Delta}, & C_{32} &= \frac{\nu_{23} + \nu_{21}\nu_{13}}{E_1 E_2 \Delta}, & C_{33} &= \frac{1 - \nu_{12}\nu_{21}}{E_1 E_2 \Delta} \\ C_{44} &= G_{12} \\ \Delta &= \frac{1 - \nu_{23}\nu_{32} - \nu_{13}\nu_{31} - \nu_{12}\nu_{21} - 2\nu_{32}\nu_{13}\nu_{21}}{E_1 E_2 E_3} \end{aligned} \quad (27)$$

where E_1 and E_2 are the transverse Young modulus (in-plane directions), E_3 is the Young modulus along with the fibers direction, ν_{ij} is the Poisson ratio characterizing the deformation rate in direction j when a force is applied in direction i , G_{ij} is the shear modulus which characterizes the variation angle between directions i and j . The elastic properties can be written as a function of the matrix components, C_{ij} ,

$$\begin{aligned}
E_1 &= \frac{\Psi}{C_{22}C_{33} - (C_{23})^2}, & E_2 &= \frac{\Psi}{C_{11}C_{33} - (C_{13})^2}, & E_3 &= \frac{\Psi}{C_{11}C_{22} - (C_{12})^2} \\
\nu_{12} &= \frac{C_{12}C_{33} - C_{13}C_{23}}{C_{22}C_{33} - (C_{23})^2}, & \nu_{13} &= \frac{C_{13}C_{22} - C_{12}C_{23}}{C_{22}C_{33} - (C_{23})^2}, & \nu_{23} &= \frac{C_{23}C_{11} - C_{12}C_{13}}{C_{11}C_{33} - (C_{13})^2} \\
G_{12} &= C_{44} \\
\Psi &= C_{11}C_{22}C_{33} + 2C_{23}C_{13}C_{12} - C_{11}(C_{23})^2 - C_{22}(C_{13})^2 - C_{33}(C_{12})^2
\end{aligned} \tag{28}$$

The straight-forward procedure to extract homogenized elastic properties of a composite material consists in the following:

- (1) Applying three distinct macroscopic deformation gradients along three different and well-defined directions of the RVE, originating three strain-states, one at a time: (1) $\varepsilon_{11} = 1$ with $\varepsilon_{22} = \gamma_{12} = 0$, (2) $\varepsilon_{22} = 1$ with $\varepsilon_{11} = \gamma_{12} = 0$; and (3) $\gamma_{12} = 1$ with $\varepsilon_{11} = \varepsilon_{22} = 0$;
- (2) For each prescribed macroscopic deformation gradient, imposing the periodic boundary conditions and solving the boundary problem – in the calculation of the RVE stiffness matrix, it is considered a different constitutive matrix, $\mathbf{c}$, for each constituent of the RVE;
- (3) Based on the displacement field obtained, computing of the stress fields at the integration points of the RVE: using $\boldsymbol{\varepsilon} = \mathbf{B} \cdot \mathbf{u}$ and $\boldsymbol{\sigma} = \mathbf{c} \boldsymbol{\varepsilon}$;
- (4) Homogenizing the stresses using the volume average of the stress fields obtained in 3 – Eq. (19)
- (5) Using the constitutive relations Eq. (27), obtaining the constants C_{ij} knowing the homogenized stress state and the correspondent prescribed strain state;
- (6) Determination of the elastic properties using the relations Eq. (29).

The previously described procedure cares in the determination of one material constant: C_{33} . The longitudinal Young modulus, E_3 , is then approximated using the rule of mixtures (ROM) (Pan *et al.*, 2016), which permits to calculate C_{33} and, consequentially, the quantity Ψ :

$$E_3^* = V_{\%f} E_f + (1 - V_{\%f}) E_m \tag{29}$$

being $V_{\%f}$ the volume fraction of fibers in the composite laminate.

Numerical Solutions

In this section, the NNRPIM is combined with the homogenization technique to yield effective elastic properties of a composite material, using different RVEs.

In the NNRPIM formulation, it is considered a second-degree configuration of influence-cells, a null polynomial basis in the construction of the interpolation functions ($m = 0$), and also $c = 0.0001$ and $p = 0.9999$ as shape parameters (the optimization of the shape parameters in the NNRPIM is addressed in several studies such as (Dinis *et al.*, 2007; Moreira *et al.*, 2016; Dinis *et al.*, 2008)). The meshless solutions are also compared with FEM solutions using the same level of discretization (an equal number of nodes discretizing the RVE).

Throughout the numerical examples, a composite material is fixed, having a unidirectional graphite fiber (AS4) as the reinforcement phase and epoxy (3501–6) as the matrix phase. The mechanical properties of the fibers are: $E_3 = 235$ GPa, $E_2 = 14$ GPa, $G_{12} = 28$ GPa, $\nu_{32} = 0.20$ and $\nu_{21} = 0.25$. The elastic properties of the matrix are: $E_3 = 4.8$ GPa, $E_2 = 4.8$ GPa, $G_{12} = 1.8$ GPa, $\nu_{32} = 0.34$ and $\nu_{21} = 0.34$.

To represent the fiber composite material, three different RVEs were considered in this work: two simplified periodic RVEs (called Type 1 and Type 2) and an additional RVE found in the literature (Ghayoor *et al.*, 2018) with a random distribution of fibers (Type 3) – Fig. 3. As previously mentioned, in the computation of the stiffness matrix of the RVE domain, one of two

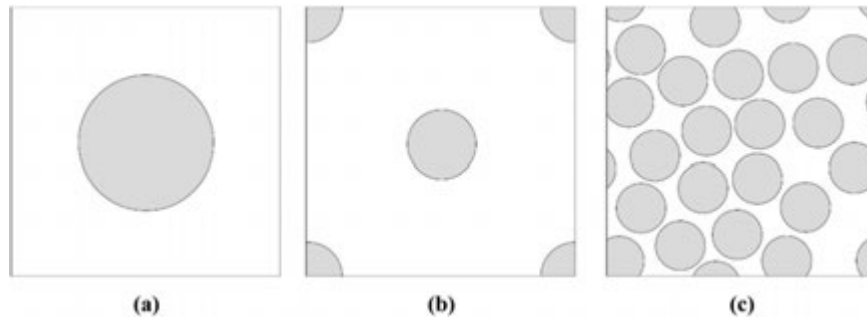


Fig. 3 (a) Type 1 RVE; (b) Type 2 RVE; (c) Type 3 RVE.

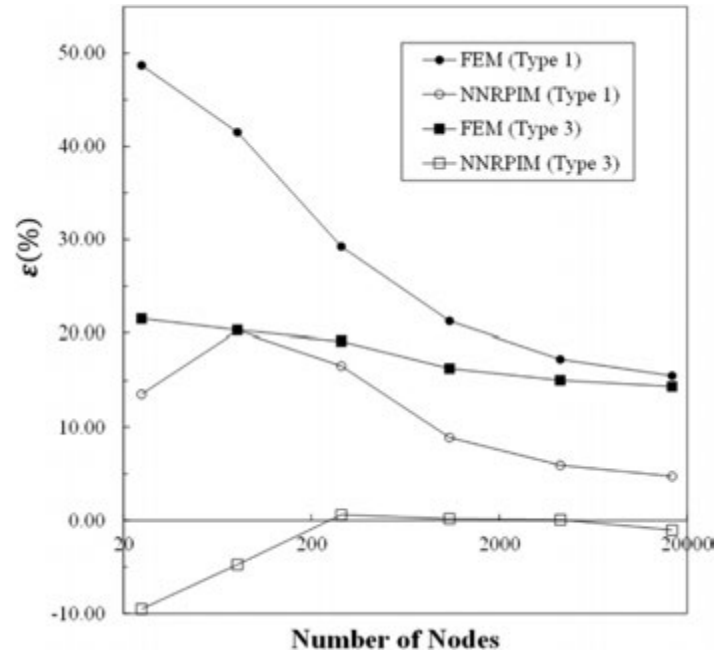


Fig. 4 Convergence studies for the FEM and NNRPIM using two configurations of RVEs. The percentage error is calculated based on the present numerical solution and uses the Hashin and Rosen solution as a reference. Reproduced from Hashin, Z., Rosen, B.W., 1964. The elastic moduli of fiber-reinforced materials. *J. Appl. Mech.* 31 (2), 223–232. Available at: <https://doi.org/10.1115/1.3629590>.

constitutive matrixes were assigned to the integration points, depending on if the integration point is within the fiber domain or the matrix domain.

Convergence Study

Type 1 and Type 3 RVE configurations (Fig. 3) are used to perform convergence studies on the FEM and the NNRPIM, using a fiber volume fraction of $V_{vf} = 0.6$ (which means that the area of the circles represented in the RVE occupies 60% of the full-square RVE area). The graphs of Fig. 4 represent the variation of the percentage error in the calculation of the transverse Young modulus as a function of the number of nodes used in the analysis (considering regular nodal meshes). The mentioned error is calculated based on the Hashin and Rosen (1964) solution which, for the given fiber volume fraction, considers $E_1 = 9.40 \text{ GPa}$. The error is calculated using the following expression:

$$\varepsilon(\%) = \frac{E_1|_{\text{Hashin and Rosen}} - E_1|_{\text{present}}}{E_1|_{\text{Hashin and Rosen}}} \times 100 \quad (30)$$

From Fig. 4, it can be concluded that the NNRPIM has faster convergence rates than the FEM. Additionally, the NNRPIM is capable to deliver, in all cases, lower percentage errors than the FEM, using exactly the same nodal mesh. Thus, this convergence study corroborates two of the advantages of the NNRPIM over the FEM that are often mentioned in the literature (Belinha, 2014): the higher accuracy and the faster convergence.

Furthermore, for the RVE with a random distribution of fibers, the errors obtained can be even lower than for RVEs with periodic geometries: both FEM and NNRPIM curves related to Type 3 RVE are placed below the curves concerning Type 1 RVE.

Despite the different convergence rates, a regular nodal mesh with 65×65 nodes (4356 nodes in total) was selected for further analysis.

Benchmark Example

The validation of the present meshless approach was performed through a comparison between the effective elastic properties obtained with the NNRPIM with FEM, literature solutions and also analytical methods, such as the rule of mixtures (ROM) and the semi-empirical model of Halpin-Tsai (Halpin and Kardos, 1976). To obtain the rule of mixtures' solutions, the following expressions are used (Pan et al., 2016):

$$\begin{aligned}
E_3 &= V_{\%f} \cdot E_f + (1 - V_{\%f}) \cdot E_m \\
E_1 = E_2 &= \frac{E_f \cdot E_m}{E_m \cdot V_{\%f} + E_f \cdot (1 - V_{\%f})} \\
\frac{1}{G_{21}} &= \frac{V_{\%f}}{G_f} + \frac{(1 - V_{\%f})}{G_m} \\
v_{32} &= V_{\%f} \cdot v_f + (1 - V_{\%f}) \cdot v_m \\
v_{21} &= \frac{v_f \cdot v_m}{v_m \cdot V_{\%f} + v_f \cdot (1 - V_{\%f})}
\end{aligned} \tag{31}$$

where the subscript $\langle \cdot \rangle_f$ represents the respective elastic constant of the fiber and $\langle \cdot \rangle_m$ represents the corresponding elastic constant of the matrix. As for the Halpin-Tsai model, the following expressions are used (Mallick, 2007):

$$\begin{aligned}
E_1 &= \frac{1 + 2\eta V_{\%f}}{1 - \eta V_{\%f}} E_m \\
E_2 &= \frac{1 + 2\eta V_{\%f}}{1 - \eta V_{\%f}} E_m \\
E_3 &= V_{\%f} \cdot E_f + (1 - V_{\%f}) \cdot E_m \\
G_{21} &= \frac{1 + 2\eta V_{\%f}}{1 - \eta V_{\%f}} G_m \\
v_{32} &= V_{\%f} \cdot v_f + (1 - V_{\%f}) \cdot v_m \\
v_{21} &= \frac{1 + 2\eta V_{\%f}}{1 - \eta V_{\%f}} v_m
\end{aligned} \tag{32}$$

where the quantity η is given by $\eta = [(p_f/p_m) - 1]/[(p_f/p_m) + \xi]$, depending on the elastic property that is being calculated. p_f is the corresponding elastic property of the fiber, p_m the same elastic property regarding the matrix and ξ is a measure of the reinforcement geometry, packing geometry, and loading conditions ($\xi = 2$ in all cases of this work since, in the determination of E_1 , E_2 , G_{21} and v_{21} for square geometries with a larger longitudinal length (Mallick, 2007), ξ assumes that value). For example, if it is intended to establish E_1 , using the Halpin-Tsai model:

$$E_1 = \frac{1 + 2\eta|_{E_1} V_{\%f}}{1 - \eta|_{E_1} V_{\%f}} E_m \text{ with } \eta|_{E_1} = \frac{(E_{1f}/E_{1m}) - 1}{(E_{1f}/E_{1m}) + 2} \tag{33}$$

Considering the same material used in the previous section, with the same volume fraction of fibers ($V_{\%f} = 0.6$), the solutions presented in Table 1 are obtained using the three configurations of RVEs.

As can be seen in Table 1, the numerical solutions are within the same range of the literature and empirical laws' solutions, proving the validity of the obtained results. Analyzing those solutions, it is observable that the NNRPIM achieves more accurate solutions than the FEM, with the latter tending to overestimate the transverse Young modulus and the shear modulus. Additionally, it is also visible that Type 2 RVE underestimates transverse Young modulus and the shear modulus for both numerical methods.

Table 1 Effective elastic properties for the composite material AS4/3501-6 (with fiber volume fraction = 0.6), considering various sources

		E_3 (GPa)	E_2 (GPa)	v_{32}	v_{21}	G_{21} (GPa)
Empirical laws	ROM	142.92	7.93	0.26	0.28	3.57
	Halpin -Tsai	142.92	9.2	0.26	0.28	3.57
Literature	C. T. Sun, R. S. Vaidya [Type 1] (Sun and Vaidya, 1996)	142.6	9.6	0.25	0.35	3.1
	C. T. Sun, R. S. Vaidya [Type 2] (Sun and Vaidya, 1996)	142.6	9.2	0.25	0.38	3.35
	Hashin, Z., Rosen, B. W. Hashin and Rosen (1964) ^a	142.9	9.40 (9.10)	0.25	0.39 (0.37)	3.42 (3.26)
	J.-W. Lee, I.M. Daniel - Experiment 1 (Lee and Daniel, 1990)	142	10.3	—	—	3.8
	C.T. Sun, S.G. Zhou - Experiment 2 (Sun and Zhou, 1988)	139	9.85	0.3	—	—
	FEM [Type 1]	142.92	11.02	0.24	0.30	4.23
Numerical solutions	FEM [Type 2]	142.92	10.66	0.24	0.31	4.06
	FEM [Type 3]	142.92	10.81	0.24	0.31	4.13
	NNRPIM [Type 1]	142.92	9.96	0.22	0.28	3.88
	NNRPIM [Type 2]	142.92	8.83	0.240	0.31	3.36
	NNRPIM [Type 3]	142.92	9.41	0.240	0.30	3.56

^aValues in parentheses indicate lower bounds.

Effect of Volume Fraction of Fibers on Homogenized Elastic Properties

For periodic RVEs (Type 1 and Type 2), the maximum volume fraction of fibers can be obtained using:

$$\%V_f|_{\max} = \frac{\pi(L/2)^2}{L \times D} = \frac{\pi}{4} \simeq 0.785 \quad (34)$$

where $L = D$ are the dimensions of the square RVE. For the Type 3 RVE, due to the random distribution of fibers with the same radius, the maximum value of volume fraction of fibers is about 0.5. Therefore, having these maximum values in mind, two effective elastic properties of the AS4/3501-6 composite are computed as a function of the fiber volume fraction. The graphs of Fig. 5 show the NNRPIM and FEM solutions, but also the solutions provided by the rule of mixtures (ROM) and the Halpin-Tsai (H-T) model for comparison purposes.

As expected, it is clear from the graphs that the transverse modulus increases (non-linearly) with the increase of the fiber volume fraction. The graphs created using the FEM (concerning the three types of RVEs) are very similar between each other but are further away from the same representations obtained with the analytical methods. As regards the NNRPIM curves, they are closer to the analytical models than the FEM curves. As for the in-plane shear modulus, similar remarks can be made: there is a good agreement between the numerical and analytical solutions, and the NNRPIM curves are closer to the ROM and H-T curves than the FEM ones.

If one takes a closer look to Fig. 5, it can be also seen that the analysis considering a RVE with a random distribution of fibers (Type 3) provides closer curves to the H-T curve when compared with the periodic and simplified models (Type 1 and Type 2).

Effect of the RVE Size

To verify if the effective elastic properties are dependent on the RVE size, the transverse Young modulus is calculated using two RVE configurations (Type 2 – periodic - and Type 3 - random), two fiber volume fractions ($V_{\%f} = 0.4$ and $V_{\%f} = 0.6$) and the two numerical methods (NNRPIM and FEM), with the increase of the characteristic length of the RVE side – as represented in Fig. 6.

The solutions for the transverse Young modulus computed with the RVEs with size 1, 2 and 3 are represented in Fig. 7:

As Fig. 7 highlights, it seems that the material property does not strongly depend on the characteristic size of the RVE for a periodic distribution of fibers. This means that the consideration of a smaller RVE does not introduce perturbations in the

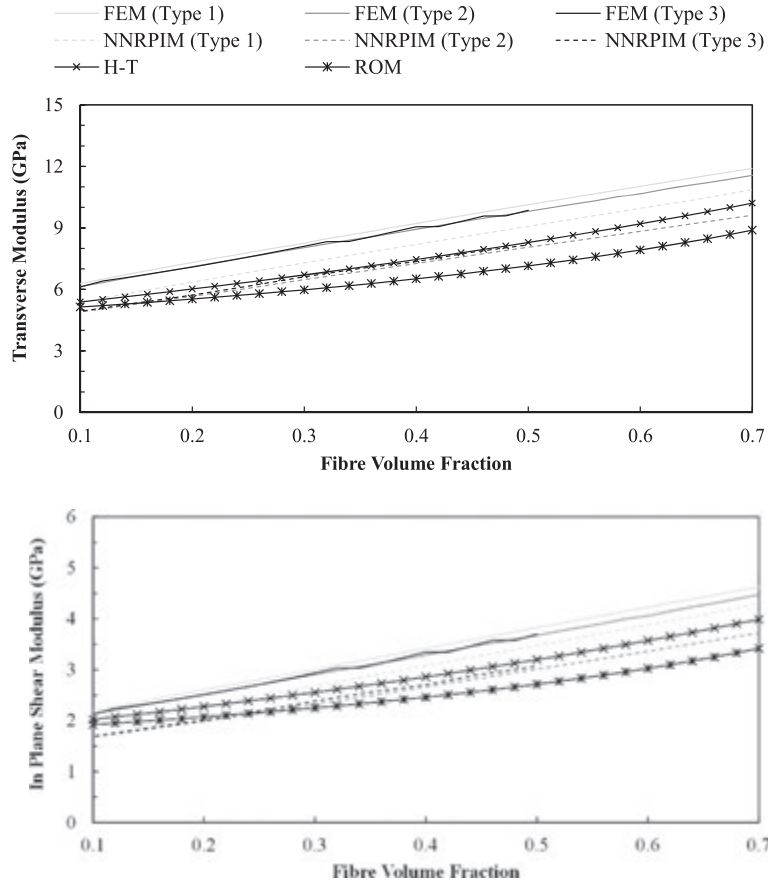


Fig. 5 Effective elastic properties as a function of the fiber volume fraction.

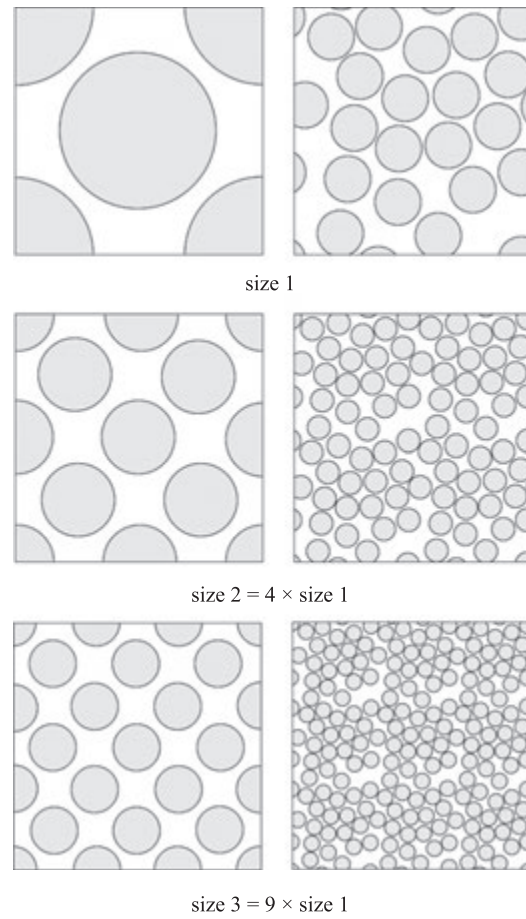


Fig. 6 Two RVE configurations with three sizes.

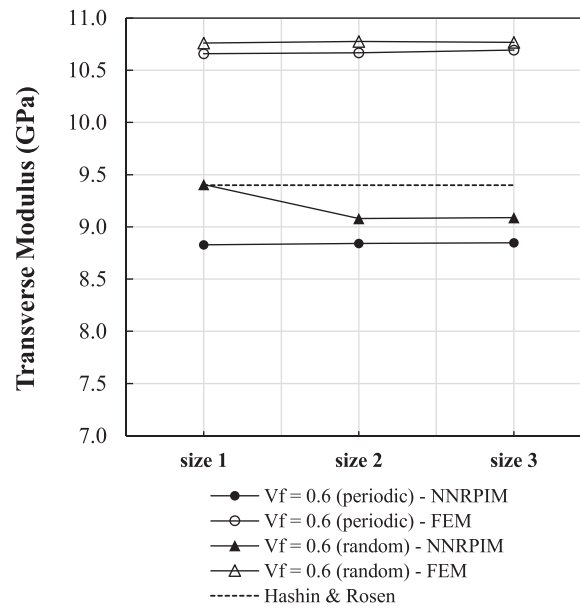


Fig. 7 Influence of the RVE size on the effective elastic properties.

computation of the effective elastic properties, for both the FEM and the NNRPIM. In the case of the RVE with a random distribution of fibers, Fig. 7 shows a small decrease of the transverse Young modulus with the size of RVE when the NNRPIM is used. This decrease cannot be seen as significant since the difference between the solution for size 1 and size 3 is less than 4.5%. Therefore, from the numerical results computed in this work, the size of the RVE does not significantly influence the homogenized elastic properties.

Conclusions

In this work, the Natural Neighbor Radial Point Interpolation Method was used to homogenize elastic properties of a composite material. Using Representative Volume Elements (RVEs), and applying a scale transition technique, the heterogeneous microstructure of a fiber composite found mechanically equivalence in a homogeneous medium. The micromechanical approach used in this work considered the composite laminates as two-dimensional solids in plane-strain state.

The MATLAB® algorithm developed, combining the NNRPIM formulation with the homogenization technique, was validated through several numerical examples. The developed computational tool is capable to compute effective elastic properties of a heterogeneous material knowing its microstructure and the elastic properties of its constituents.

Despite the higher computational cost of the NNRPIM when compared with the FEM (for the same level of discretization), the meshless approach achieved faster convergences and more accurate effective elastic properties, when a comparison is made between the NNRPIM and FEM solutions with some solutions provided in the literature. Thus, the advantage of using the NNRPIM over the FEM, for material homogenization problems, was proved.

In general, the effect of the fiber volume fraction on the effective elastic properties was well captured by the NNRPIM, being the solutions computed with the NNRPIM for several fiber volume fractions closer to the ones predicted by analytical laws such as the rule of the mixtures and the Halpin-Tsai semiempirical model. Unexpectedly, in this work, the obtained numerical results shown that the effective elastic properties did not depend on the size of the RVE.

Acknowledgments

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Design Criteria for Pultruded Structural Elements

Fausto Tucci¹, University of Salerno, Fisciano, Italy

Alexander Vedernikov¹, Skolkovo Institute of Science and Technology, Moscow, Russia

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Introduction

In the last decades, structures made of traditional materials have faced the problems of increased restoration and maintenance costs due to their relatively short service-life. This issues promoted the introduction of composites, which are less prone to corrosion (Martins *et al.*, 2017), (Smith *et al.*, 1998), (Back and Will, 2008), lighter (Nguyen *et al.*, 2015), (Bai and Yang, 2013), and easier to build (Mottram and Zheng, 1996), to the construction sector.

FRPs have been widely employed as structural element, whereas high strength to weight ratio is pursued (Bank, 2007). These materials consist of two main constituents: dispersed fibrous reinforcement (providing mechanical resistance) and low-density matrix (ensuring the corrosion resistance) embedding the fibers. The matrix is usually constituted by a polymeric resin mixed with other components aimed to improve the resin flow and reaction during processing, to provide the composite with the required physical properties (Devendra and Rangaswamy, 2013).

Among all the FRPs, the composites manufactured by pultrusion process cover a particular role due to the marked anisotropy and the high content of fibrous reinforcement achievable (Starr, 2000). The pultrusion is a continuous automated process for the production of FRP profiles with constant cross-section. The pultruded composites are widely applied as structural elements in many different fields: civil, marine, automotive, aeronautic, energy and bridge construction (Vedernikov *et al.*, 2020a,b,c). Pultruded profiles have been employed in the construction of buildings and civil structures as beams (Ascione and Mancusi, 2013), columns (Xie *et al.*, 2019), reinforcing rebars (Benmokran *et al.*, 1995), for the rehabilitation or retrofitting of existing concrete structures (Altaee *et al.*, 2017).

Generally, the pultruded profiles present a marked anisotropy, depending on the fibrous architecture. In the most of the cases, the pultruded profiles are reinforced with longitudinal unidirectional fiber rovings. Fabric or mat reinforced layers are often used to provide the pultruded surface with higher resistance to tangential transverse loads (Bank, 2007). Due to this kind of reinforcement architecture, pultruded composites exhibit their best mechanical performances in the longitudinal direction (Haj-Ali and Kilic, 2002).

The structural performance of the pultruded elements is strictly related to the process parameters employed during the production stage. Once defined the cross-section shape and the profile length, the main operative parameters tunable in pultrusion processes are the pulling velocity, the heating platens temperature and, in case of injection pultrusion, the resin pressure at the injection slots (Baran, 2015). The heating temperature and the pulling speed define the curing cycle provided to the polymeric system to achieve the polymerization reaction. Incomplete reaction at the die outlet provokes shape distortions, delamination and internal defects (Vedernikov *et al.*, 2020a,b,c). On the other hand, excessive thermal energy also results in low-quality profiles due to possible local or global degradation of the matrix. The contact between advancing material and the pultrusion die generates forces resisting to the pulling action, which produce internal stresses in the pultruded products (Safonov *et al.*, 2018).

The pultruded FRP structures should also be designed by taking into account circumstances that can influence their durability and design life: the chemical–physical environment where the structure is applied (including UV (Carra and Carvelli, 2014; Stazi *et al.*, 2015), temperature influences (Ghadimi *et al.*, 2017), (Russo *et al.*, 2016), (Turvey and Sana, 2016), humidity, water (Xin *et al.*, 2017), (Cabral-Fonseca *et al.*, 2012), (Gómez *et al.*, 2012), (Kafodya *et al.*, 2015) and chemical agents (Gómez *et al.*, 2012)); time-dependent influences such as creep and wear; fatigue; accidental loads including: (fire, lightning strike, impact (Guades and Aravinthan, 2013; Guades *et al.*, 2013), explosion); the transportation and installation phases; as well as inspection and maintenance. A further important factor, remarkably affecting the mechanical performances of pultruded structures, is related to the connections. Indeed, pultruded FRPs present particular issues at the bolted joints, which should be taken into account in the design step (Feroldi and Russo, 2017), (Feroldi and Russo, 2016), (Russo, 2019).

Design Criteria for Pultruded Structural Elements

“Allowable Stress Design” (ASD), “Load and Resistance Factor Design” (LRFD) and “Limit State” are the three main ideologies currently employed in structural design of elements produced by pultrusion process.

According to ASD philosophy described by Eq. (1), under nominal service load no element within the structure should reach its ultimate stress:

$$\sigma_{reqd} \leq \frac{\sigma_{ult}}{SF}, \quad (1)$$

¹The two authors contributed equally to the development of this work.

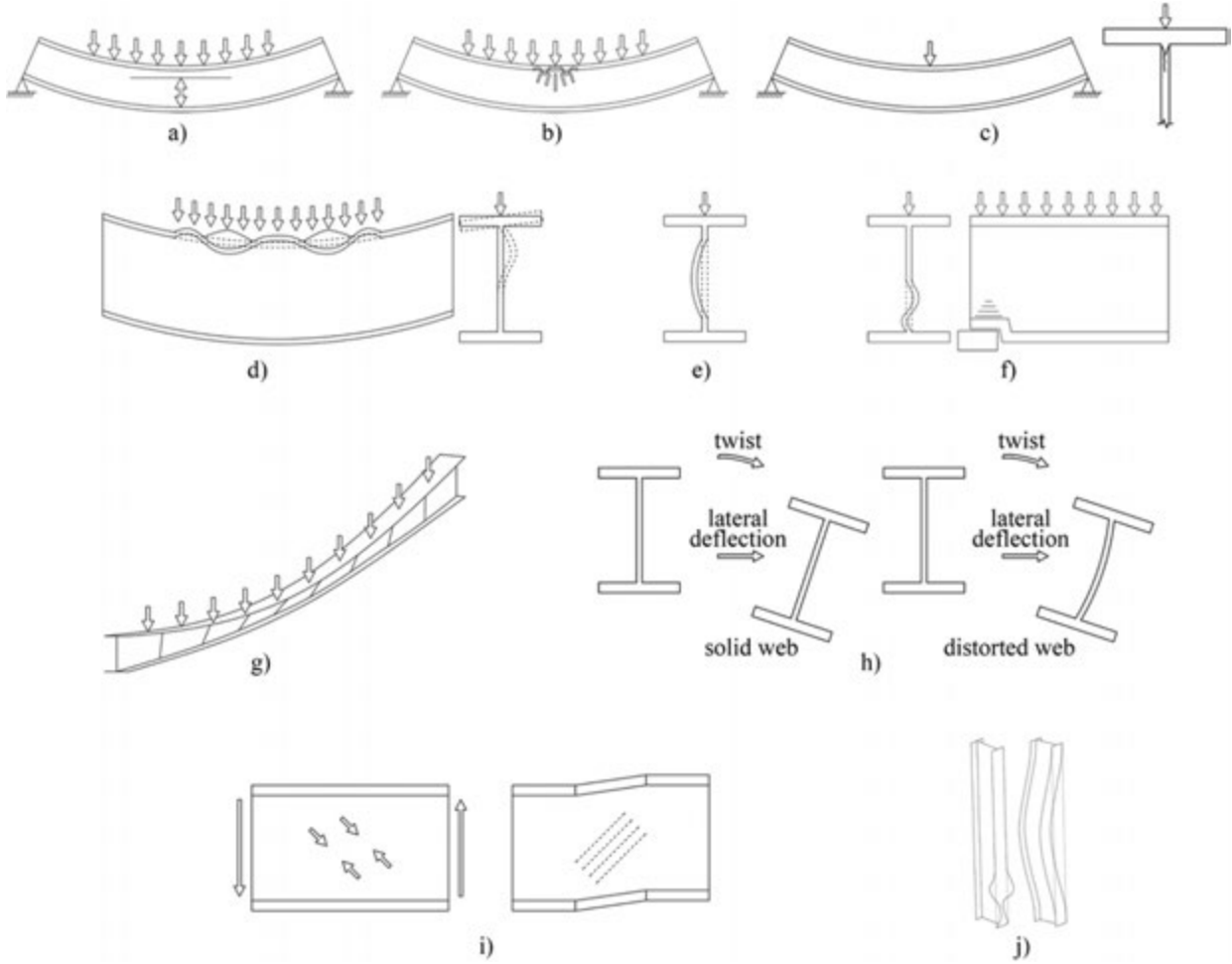


Fig. 1 Failure modes of composite elements. (a) Failing of SLS requirements due to excessive deflection during flexural loading; (b) beam failure due to flexural loading; (c) beam failure due to shear loading; (d) compression flange local buckling due to flexural loading: front view and side view of the profile; (e) web local buckling due flexural loading. Side view of the profile; (f) web transverse crushing failure: side view and front view of the profile; (g) lateral torsional buckling failure; (h) buckling failure: lateral torsional buckling mode (side view of the profile) and lateral-distortional buckling mode (side view of the profile) respectively; (i) web local buckling due shear loading. Front view of the profile; (j) beam failure due to buckling: local buckling mode and global (Euler) buckling mode respectively.

where σ_{reqd} represents the required stress (or design stress in every member), σ_{ult} is the ultimate strength of the material and SF represents safety factor. While being over-conservative in some cases, this approach provides underestimated results in others as loads are considered at service values only. As the Serviceability Limits are not considered in ASD, loading of a structure can result in a failure to meet service requirements, even if the element is still safe from the bearing capacity point of view. This structural design philosophy is widely accepted in manuals of pultrusion companies in the United States (Bank, 2007).

LRFD philosophy, on the other hand, is represented by Eq. (2). The main idea of this approach is that applied loads are increased according to their combinations and types, while resistance of the structural members is decreased due to material properties variability, type of resistance required etc. A number of coefficients are utilized for the mentioned scaling procedure. Moreover, the serviceability limit state is considered here:

$$R_{reqd} \leq \phi R_n, \quad (2)$$

where R_{reqd} is the required resistance of a structural member, R_n represents the nominal resistance of structure or material and ϕ is the resistance factors.

Finally, the Limit State design philosophy is based on the assumption that a structure is considered failed whereas it is not capable anymore to perform the function for which it has been designed. A deeper analysis of this philosophy is provided in the following section.

Number of factors employed in each philosophy creates a critical difference between them. One factor for resistance and several factors for loads are used within LRFD, while ASD utilizes only a safety factor. LRFD, being more advanced and rational, is applied more often than ASD.

Having appeared first in Norway in 1963, the Performance-Based Design (PBD) philosophy is spreading across the whole world in the last decades (Inokuma, 2002). At the core of PBD philosophy are the structural objectives that have to be fulfilled. These goals are initially set by engineers and client together. The final design must comply with the predetermined targets. This philosophy has not found its place in structural design of pultruded FRPs. Careful research and understanding of possible applications of this philosophy can be another target for the pultrusion society to go.

This article reviews analytical, experimental, and numerical investigations that have been done so far regarding the structural behavior of pultruded profiles. Certain analytical formulas available in the literature for the design of FRP members at both ultimate and serviceability limit state are presented as well.

Limit States

The design process of FRP structures is similar to that of steel, with the only regards of the orthotropic and linear elastic response of composites to be considered (Correia *et al.*, 2010). The serviceability limit states (SLS) and the ultimate limit states (ULS) check should be performed for the elements and connections. The limit state can be defined as the case when the structure ceases to fulfill its intended purpose in some way.

To examine the condition in which none of the limit states is violated, the partial factors method should be applied, keeping in mind all the values of actions and resistances acting on the structure. The corresponding requirement must be ensured (Grimaldi, 2007):

$$E_d < R_d = \varphi L_d, \quad (3)$$

where E_d represents the design value of the action, R_d is the related capacity (resistance or deformation) within a considered limit state, φ is a corrective safety factor accounting for uncertainties of service conditions and L_d is the actual property limit of the designed element. By employing appropriate partial factors for the corresponding limit state, the design value can be calculated.

Serviceability Limit State

In case the structural behavior falls below the prescribed service requirements, the SLS is violated (American Society for Civil Engineers, 2010). The SLS to be considered are the following:

- (1) deformations or deflections leading to an inappropriate appearance of the structure as well as its ineffective use (including disruption of machines or service processes) either to damage of finishes or nonload bearing elements;
- (2) vibrations generating discomfort to staff, damage of the building or its indoor elements, or confining limitation on its effective use;
- (3) cracking or delamination of the FRP composites, which is likely to promote the worsening of the appearance, durability, or waterproof properties;
- (4) local damage of FRP composite (due to impact or local bearing failure) experiencing extreme stress, which likely causes loss of durability. Linear elastic theory should be the basis for the SLS calculations (Clarke, 1996).

Fig. 1 depicts the main failure modes of pultruded I-beams.

There is no difference whether a material or structural level is considered, the bending behavior of FRP elements demonstrates certain distinctions compared to the conventional material design. Structural elements made of FRP possess relatively low elastic modulus in contrast to steel (Ascione *et al.*, 2015a,b), and therefore bending stiffness as well. As a result, in the case of FRP profiles, the design process has its peculiarities and is driven either by SLS, restricting deflections (Satasivam and Bai, 2014; Minghini *et al.*, 2016) (Fig. 1(a)), or by a buckling phenomenon (considering thin-walled sections) opposing to the ULS (Barros da *et al.*, 2007). This is the reason why pultruded profiles are frequently applied in the cases of lighter and shorter spans.

Barbero *et al.* proved that in bending pultruded beams possess linear-elastic behavior even for large deflections. It can be explained by huge elongations allowed by fibers (4%) and matrix (4.5%). This value is certainly higher than those for conventional materials such as concrete (cracking) and steel (plasticity) (Barbero *et al.*, 1991).

Considering the shear contribution to the overall deformation, in other words relying on analytical beam models based upon Timoshenko theory, flexural deflections of pultruded profiles can be calculated and, therefore, the corresponding SLS criteria can be verified. Indeed, the shear contribution can be essential and should be included in the overall consideration. Long-term deformations of composites have their own peculiarities as a result of different viscoelastic behavior and polymeric properties of the matrix (Correia *et al.*, 2010).

Ultimate Limit State

ULS is the case when a structure reaches its critical point, and further growth of the load will lead to a collapse or another form of failure. It may place both inhabitants and performance of the structural components in danger. The ULS cases to be considered are the following: violation of the construction's equilibrium or even any part of this structure accounted as a rigid body; collapse due

to exceeding deformation, break, or stability loss of the structure in general or any of its parts in particular (Clarke, 1996). Historically, in the related documents, design codes, and guidelines, more attention was paid to ULS than to SLS owing to its importance, ensuring the buildings' safety from the first glance (American Society for Civil Engineers, 2010).

Pultruded FRP members can fail owing to several reasons that must be considered during ULS design. This means that the design resistance should be not less than the action or resistance force acting on the structure. The following states are to be verified: (1) flexural strength; (2) shear strength (3) web local buckling due to flexure; (4) web local buckling due to shear; (5) web local buckling due to flexure and shear; (6) compression flange local buckling; (7) lateral–torsional buckling; (8) web transverse crushing; (9) axial strength.

Flexural strength

The design value of the internal bending moment (M_{sd}) in any cross-section of the beam shall satisfy (Clarke, 1996):

$$M_{sd} \leq M_{Rd}, \quad (4)$$

where M_{Rd} is the design moment resistance of the cross-section corresponding to the flexural failure of FRP member. Its value can be computed as follows (Correia *et al.*, 2010):

$$M_{Rd} = \sigma_{x,u} \cdot W_x, \quad (5)$$

where $\sigma_{x,u}$ is the longitudinal failure stress (either compressive or tensile) of the pultruded profile and W_x is the cross-sectional elastic modulus about the strong axis.

Typical failure occurring because of bending moment is shown in the Fig. 1(b).

Shear strength

In contrast to the tensile strength, composite-based profiles possess a relatively low value of shear strength. This fact should be considered during the design stage, because shear deformation is significant and the effects can be crucial (Barros da *et al.*, 2007). Nonuniformity in load application or cross-section can cause large shear stresses and eventually lead to sudden shear failure (Davalos *et al.*, 2002).

The design value of the shear force (V_{sd}) in any section of the beam should be less than the critical value (Clarke, 1996):

$$V_{sd} \leq V_{Rd} \quad (6)$$

In the case of thin-walled open sections the critical shear force (V_{Rd}) of a FRP beam under bending can be calculated as follows (Correia *et al.*, 2010):

$$V_{Rd} = \frac{\tau_u \cdot I_x \cdot t}{S_x} \approx \tau_u \cdot A_v, \quad (7)$$

where τ_u represents the in-plane shear strength of the beam, S_x is the first moment of area about the strong axis, t represents the laminate (web/flanges) thickness and A_v is shear area (for most common element it is equal to the web(s) of the element) (Correia *et al.*, 2010).

To proceed with the design of the elements, corresponding mechanical properties are needed. Conducting an experiment in which only pure shear is applied toward pultruded FRP materials is a challenging task. Currently adopted standard test methods have technical weaknesses. For instance, an accurately manufactured coupon of pultruded material is needed, as well as expensive and precise experimental tooling.

Typical failure occurring due to the action of shear forces is shown in the Fig. 1(c).

Buckling

As it was mentioned before, pultruded elements exhibit orthotropic behavior. A composite's strength and elastic constants are much higher in the longitudinal direction compared to the transverse directions (Pecce and Cosenza, 2000). As a consequence, designers should consider pultruded structures as the elements being linear, elastic, homogeneous, and transversely isotropic with the isotropy plane perpendicular to the fiber direction (Ascione *et al.*, 2011). The bearing capacity of the pultruded elements generally defined by the critical buckling load. This is due to the fact that FRP composites being loaded, first of all, violate the deformability and stability requirements and not the strength requirements (Ascione and Mancusi, 2013).

As far as steel has different mechanical properties compared to composites, its design guides regarding buckling calculations cannot be applied toward pultruded elements. Moreover, orthotropic behavior creates some peculiarities pushing forward local buckling issues of section components, flanges, and web (Pecce and Cosenza, 2000).

Slender elements are generally subjected first to global (Euler) buckling failure rather any other types. This is in stark contrast to the behavior of short columns, where local buckling is more likely to occur before global buckling. As a consequence, local buckling leads to large deformations resulting in global buckling or material degradation (crippling phenomenon).

The majority of the theoretical models developed so far have focused on the prediction of FRP structural elements' behavior simply transferring formulations of isotropic models to the anisotropic ones. The results of contemporary analytical and FEM methods correlate well with data obtained during the experiments of both long and short columns (Barbero and Tomblin, 1994).

All members shall be checked for local buckling of their flange(s) and web(s) that are subjected to compressive stresses and shear stresses due to flexure of the member. These cases are described in detail in the following subarticles with respect to the most

Table 1 Different approaches and their features toward investigation of pultrusion elements during buckling loading and buckling failure

Approaches		References
Type of buckling	Global (Euler)	(Barbero and Tomblin, 1994), (Puente <i>et al.</i> , 2006), (Harries <i>et al.</i> , 2017), (Gan <i>et al.</i> , 1999a,b), (Hashem and Yuan, 2001), (Di Tommaso and Russo, 2003)
	Local	(Barbero, 1993), (Ascione <i>et al.</i> , 2016), (Puente <i>et al.</i> , 2006), (Mottram <i>et al.</i> , 2003a,b), (Nunes <i>et al.</i> , 2013), (Bank <i>et al.</i> , 2008)
	Flexural	(Minghini <i>et al.</i> , 2008), (Roberts, 2002), (Roberts and Masri, 2003)
	Torsional	(Minghini <i>et al.</i> , 2008), (Roberts, 2002), (Roberts and Masri, 2003)
	Flexural-torsional	(Minghini <i>et al.</i> , 2008), (Pandey <i>et al.</i> , 1995), (Davalos <i>et al.</i> , 1997), (Qiao <i>et al.</i> , 2003), (De Lorenzis and La Tegola, 2005), (Shan and Qiao, 2005)
	Lateral-torsional	(Minghini <i>et al.</i> , 2008), (Pandey <i>et al.</i> , 1995), (Mottram, 1992b), (Razzaq <i>et al.</i> , 1996), (Sapkás and Kollár, 2002), (Nguyen <i>et al.</i> , 2014)
	Lateral	(Brooks and Thrvey, 1995), (Barbero and DeVivo, 1999), (Bai <i>et al.</i> , 2013), (Mancusi <i>et al.</i> , 2014), (Barbero and Raftoyiannis, 1994), (Turvey, 1996b)
	Distortional	(Silvestre and Camotim, 2003), (Barbero and Raftoyiannis, 1994)
	Lateral-distortional	(Davalos <i>et al.</i> , 1997), (Davalos and Qiao, 1997)
	Flexural-distortional	(Silvestre and Camotim, 2003)
Tested element type	Angle section profile	(Cardoso <i>et al.</i> , 2014a,b), (Ragheb, 2017)
	C section profile	(Silvestre and Camotim, 2003), (Wong and Wang, 2007), (Minghini <i>et al.</i> , 2008), (Cardoso <i>et al.</i> , 2014a,b), (Ragheb, 2017), (Shan and Qiao, 2005), (Razzaq <i>et al.</i> , 1996), (Nguyen <i>et al.</i> , 2014), (Kabir and Sherbourne, 1998), (Minghini <i>et al.</i> , 2009)
	Circular hollow section profile	(Puente <i>et al.</i> , 2006)
	Deck panel	(Gan <i>et al.</i> , 1999a,b)
	Frame	(Minghini <i>et al.</i> , 2008), (Minghini <i>et al.</i> , 2009)
	Girder	(Bai <i>et al.</i> , 2013)
	Hat section profile	(Silvestre and Camotim, 2003)
	Hollow box section profile	(Barbero, 1993), (Barbero <i>et al.</i> , 1991), (Estep <i>et al.</i> , 2016), (Barbero and Raftoyiannis, 1993), (Hashem and Yuan, 2001), (Cardoso <i>et al.</i> , 2014a,b), (Hashem and Yuan, 2000), (Qiao <i>et al.</i> , 2001)
	Hollow rectangular section profile	(Cardoso <i>et al.</i> , 2014)
	I section profile	(Barbero, 1993), (Ascione <i>et al.</i> , 2016), (Brooks and Thrvey, 1995), (Pecce and Cosenza, 2000), (Ascione <i>et al.</i> , 2011), (Barbero and Tomblin, 1993), (Gan <i>et al.</i> , 1999a,b), (Di Tommaso and Russo, 2003), (Minghini <i>et al.</i> , 2008), (Nunes <i>et al.</i> , 2013), (Laudiero, Minghini and Tullini, 2014), (Mancusi <i>et al.</i> , 2014), (Bank <i>et al.</i> , 2008), (Cardoso <i>et al.</i> , 2014a,b), (Cardoso <i>et al.</i> , 2015), (Fernandes <i>et al.</i> , 2015a,b), (Fernandes <i>et al.</i> , 2015a,b), (Ragheb, 2017), (Roberts, 2002), (Roberts and Masri, 2003), (Pandey <i>et al.</i> , 1995), (Qiao <i>et al.</i> , 2003), (De Lorenzis and La Tegola, 2005), (Mottram, 1992b), (Mottram, 1992a), (Sapkás and Kollár, 2002), (Nguyen <i>et al.</i> , 2014), (Barbero and Raftoyiannis, 1994), (Turvey, 1996a), (Minghini <i>et al.</i> , 2009), (Mancusi and Feo, 2013), (Ascione <i>et al.</i> , 2015a,b)
	Laminate	(Bai and Keller, 2011)
	Plate	(Saha <i>et al.</i> , 2004)
	Rectangular section profile	(Turvey, 1996b)
	Universal section profile	(Hashem and Yuan, 2001), (Hashem and Yuan, 2000)
	Wide Flange section profile	(Barbero <i>et al.</i> , 1991), (Barbero and Tomblin, 1994), (Barbero and Raftoyiannis, 1993), (Barbero and DeVivo, 1999), (Barbero <i>et al.</i> , 2000), (Barbero, 2000), (Lane and Mottram, 2002), (Di Tommaso and Russo, 2003), (Mottram <i>et al.</i> , 2003a,b), (Laudiero <i>et al.</i> , 2014), (Tomblin and Barbero, 1994), (Bank <i>et al.</i> , 1995), (Bank <i>et al.</i> , 1996), (Qiao <i>et al.</i> , 2001), (Mottram, 2004), (Davalos <i>et al.</i> , 1997), (Davalos and Qiao, 1997), (Qiao <i>et al.</i> , 2003), (Barbero and Raftoyiannis, 1994), (Turvey and Zhang, 2006)
	Z section profile	(Ragheb, 2017)

applied cross-sections. Nevertheless, the reader can refer to the ASCE standard for all the other cross-sections ([American Society for Civil Engineers, 2010](#)). Critical buckling stress should be adopted as the minimum of compression flange local buckling and web local buckling. **Table 1** reviews primary studies (experimental, analytical, and FEM) investigating the influence of the cross-sectional characteristics and load cases on the buckling behavior of pultruded elements at its different modes.

Web local buckling due to flexure

In the case of singly and doubly symmetric I-shaped members as well as singly symmetric channels bent about their strong axis critical buckling stress (σ_{cr}^{local}) due to compression web local buckling can be found as ([American Society for Civil Engineers, 2010](#)):

$$\sigma_{cr}^{local} = \frac{11.1\pi^2 t_w^2}{12h^2} (1.25\sqrt{E_{L,w}E_{T,w}} + E_{T,w}v_{LT} + 2G_{LT}), \quad (8)$$

where t_w represents the web thickness, h is the full height of the member, $E_{L,w}$ and $E_{T,w}$ are the characteristic elastic modulus of the web(s) respectively in longitudinal and transversal direction, v_{LT} represents the characteristic longitudinal Poisson's ratio and G_{LT} is the characteristic in-plane shear modulus.

Web local buckling due to shear

At cross-sections of the highest shear force, which occurs typically at supports and concentrated force points, the web may buckle in shear. For webs of I-members, back-to-back channels, single channels and square/rectangular box members bent about their strong axis, the critical shear buckling stress (τ_{cr}^{local}) can be evaluated as (American Society for Civil Engineers, 2010):

$$\tau_{cr}^{local} = \frac{t_w^2 k_{LT1} \sqrt{E_{L,w}(E_{T,w})^3}}{3h^2}, \quad (9)$$

in the case of $2G_{LT} + E_{T,w}v_{LT} \leq \sqrt{E_{L,w}E_{T,w}}$. Otherwise, it should be found from the following formula:

$$\tau_{cr}^{local} = \frac{k_{LT2} E_{T,w} t_w^2}{3h^2} \sqrt{v_{LT} + \frac{2G_{LT}}{E_{T,w}}}, \quad (10)$$

where t_w represents the web thickness, $k_{LT1,2}$ is the shear buckling coefficients, $E_{L,w}$ and $E_{T,w}$ are the characteristic moduli of the web respectively in longitudinal and in transversal direction, h represents the full height of the member, G_{LT} is the characteristic in-plane shear modulus; v_{LT} represents the characteristic longitudinal Poisson's ratio and A_s : shear area.

Web local buckling due to flexure and shear

In the case of high shear forces and high bending moments acting on the beam simultaneously, its web is subjected to combined in-plane shear stress (τ) and in-plane axial compressive (flexural) stress (σ) at the same time. Therefore, the critical web buckling stress is likely to be reduced. The following check should be performed (Bank, 2007):

$$\left(\frac{\sigma}{\sigma_{cr}^{local}}\right)^2 + \left(\frac{\tau}{\tau_{cr}^{local}}\right)^2 \leq 1, \quad (11)$$

or in terms of internal actions as (Bank, 2007):

$$\left(\frac{M_x}{M_{cr}^{local}}\right)^2 + \left(\frac{V_y}{V_{cr}^{local}}\right)^2 \leq 1. \quad (12)$$

Compression flange local buckling

In regular profiles, low values of the in-plane moduli and slenderness of the plate elements are the main reasons for the increased possibility to experience local buckling due to transverse loads. A load exceeding elastic buckling can lead to the failure characterized by detachment of the flange from the web of the profile. This is then followed by in-plane buckling of the web. For the most common single and doubly symmetric I-shaped members bent about their strong axis, as well as for T cross-section and back-to-back angles bent about their strong axis, the critical buckling stress (σ_{cr}^{local}) due to compression flange local buckling can be found as (American Society for Civil Engineers, 2010):

$$\sigma_{cr}^{local} = \frac{4t_f^2}{b_f^2} \left(\frac{7}{12} \sqrt{\frac{E_{L,f}E_{T,f}}{1 + 4.1\xi}} + G_{LT} \right), \quad (13)$$

where $E_{L,f}$ and $E_{T,f}$ are the characteristic moduli of the flange respectively in longitudinal and transversal directions, G_{LT} represents the characteristic in-plane shear modulus, b_f is the full width of the flange, t_f represents the thickness of the flange and ξ is the coefficient of restraint.

Lateral-torsional buckling

A loaded element should be braced against lateral displacement and rotation of the cross-section. In case this is not done, this could lead to the lateral-torsional buckling (Nguyen et al., 2015). Generally, this type of instability is more likely to occur in open-section (typically, I-shaped) elements subjected to transverse loads. At the moment of lateral-torsional buckling failure, the structure can be described by two phenomena: flanges displaced laterally (in relation to the transverse load direction), and twisted web that forces the whole beam to get out of its vertical plane (Bank, 2007).

In case of an I cross-section bent about its strong axis, the critical normal stress (σ_{cr}) due to lateral-torsional buckling shall be determined as follows (Correia et al., 2010):

$$\sigma_{cr} = \frac{C_b}{W_x} \sqrt{\frac{\pi^2 E_{L,f} I_y G_{LT} I}{(k_f L_b)^2} + \frac{\pi^4 E_L^2 I_y C_w}{(k_f L_b)^2 (k_w L_b)^2}}, \quad (14)$$

Table 2 Different approaches and their features toward investigation of pultrusion elements during axial loading and axial failure.

Approaches		References
Fiber type	Basalt	(Lu <i>et al.</i> , 2015)
	Carbon	(Creighton and Clyne, 2000), (Kwon <i>et al.</i> , 2019), (Chen and Ma, 1994)
	Glass	(Bai and Keller, 2009), (Cunningham <i>et al.</i> , 2015), (Zafari and Mottram, 2016), (Aydin, 2016), (Kim and Qian, 2017), (Liberatore <i>et al.</i> , 2018)
Resin type	Kevlar	(Chen and Ma, 1994)
	Epoxy	(Creighton and Clyne, 2000), (Lu, Xian and Li, 2015), (Kwon <i>et al.</i> , 2019)
	Isophthalic polyester	(Correia <i>et al.</i> , 2013), (Wang and Zureick, 1994), (Riebel and Keller, 2007), (Cunningham <i>et al.</i> , 2015), (Zafari and Mottram, 2016), (Kim and Qian, 2017)
	Phenolic	(Cordeiro <i>et al.</i> , 2016)
	Polyester	(Barbero <i>et al.</i> , 1999), (Turvey and Zhang, 2018), (Sigley <i>et al.</i> , 1991), (Sigley <i>et al.</i> , 1992), (Aydin, 2016), (Liberatore <i>et al.</i> , 2018)
	Polyurethane	(Chen and Ma, 1994)
	Vinylester	(Haj-Ali and Kilic, 2002), (Cordeiro <i>et al.</i> , 2016), (Barbero <i>et al.</i> , 1999), (Sonti and Barbero, 1996), (Mottram, 2011)
Investigation type	Analytical	(Barbero <i>et al.</i> , 1999), (Sonti and Barbero, 1996), (Correia <i>et al.</i> , 2013), (Sigley <i>et al.</i> , 1992)
	Experimental	(Turvey and Zhang, 2018), (Mottram, 2011), (Sigley <i>et al.</i> , 1991), (Sigley <i>et al.</i> , 1992), (Wang and Zureick, 1994), (Gosling and Saribiyik, 2003), (Masran <i>et al.</i> , 2013)
Tested element type	Numerical	(Creighton and Clyne, 2000), (Haj-Ali and Kilic, 2002), (Gosling and Saribiyik, 2003), (Girão Coelho <i>et al.</i> , 2015)
	Circular hollow section	(Bai and Keller, 2009)
	I section profile	(Correia <i>et al.</i> , 2013)
	Hollow box section profile	(Barbero <i>et al.</i> , 1999), (Aydin, 2016), (Kim and Qian, 2017), (Liberatore <i>et al.</i> , 2018), (Masran <i>et al.</i> , 2013)
	Plate	(Saha <i>et al.</i> , 2004), (Girão Coelho <i>et al.</i> , 2015)
	Rope	(Kwon <i>et al.</i> , 2019)
	Specimen	(Creighton and Clyne, 2000), (Haj-Ali and Kilic, 2002), (Barbero <i>et al.</i> , 1999), (Correia <i>et al.</i> , 2013), (Chen and Ma, 1994), (Mottram, 2011)
Loading/test type	Wide Flange section profile	(Barbero <i>et al.</i> , 1999)
	Compression	(Creighton and Clyne, 2000), (Haj-Ali and Kilic, 2002), (Barbero <i>et al.</i> , 1999), (Correia <i>et al.</i> , 2013), (Turvey and Zhang, 2018), (Saha <i>et al.</i> , 2004)
	Tension	(Haj-Ali and Kilic, 2002), (Lu <i>et al.</i> , 2015), (Cordeiro <i>et al.</i> , 2016), (Sonti and Barbero, 1996), (Correia <i>et al.</i> , 2013), (Kwon <i>et al.</i> , 2019)

where C_b represents the coefficient accounting for moment variation along the beam length, C_w is the warping constant, W_x is the section modulus of the strong axis, $E_{L,f}$ and G_{LT} are respectively the characteristic longitudinal modulus of the flange and the characteristic in-plane shear modulus. I_y represents the moment of inertia about the weak axis of bending, J is the torsional constant, k_f and k_w are the effective length coefficients respectively for flexural buckling of the weak axis and for torsional buckling of the section and L_b represents unbraced length of the beam.

Web transverse crushing

Points where concentrated loads or reactions meet a pultruded structural element are the weakest places of the beam in terms of web resistance to transverse forces. This occurs because webs of composite beams are especially subjected to local failure at these points, as the beam's web possess relatively low compressive strength and stiffness in the transverse direction.

Considering a design against web transverse crushing, transverse compressive strength ($\sigma_{T,c}$) of the web should be equal to critical crushing stress ($(\sigma_y)_{cr}^{crush}$) (Bank, 2007):

$$(\sigma_y)_{cr}^{crush} = \sigma_{T,c} \quad (15)$$

and the critical crushing force (F_{cr}^{crush}) is (Bank, 2007):

$$F_{cr}^{crush} = (\sigma_y)_{cr}^{crush} A_{eff}. \quad (16)$$

The effective area (A_{eff}) can be evaluated taking into account that the transverse concentrate loads and the consequent reactions at the supports produce a compressive load at the web. Therefore A_{eff} can be estimated as a function of the effective bearing length (L_{eff}), the web thickness (t_w), the flange thickness (t_f) and, if it is present, the thickness of a bearing element (t_{bp}), as described either in Eq. (17) for the case when flanges outstand on the both sides of the web (Bank, 2007):

$$A_{eff} = (t_w + 2t_f + 2t_{bp})L_{eff}, \quad (17)$$

Table 3 Design guidance and standards of pultruded structural profiles

Approaches		References
Country	UK	(CIRIA C779, 2018), (Clarke, 1996), (The Highways Agency, 2005), (Arya <i>et al.</i> , 2004), (TR57 Strengthening Concrete Structures with Fiber Composite Materials: Acceptance, Inspection and Monitoring, 2003), (BS EN 13706–1:2002. 'Reinforced plastic composites - Specification for pultruded profiles - Part 1: Designation,' British Standards Institution, 2002), (BS EN 13706–2:2002. 'Reinforced plastic composites - Specification for pultruded profiles - Part 2: Methods of test and general requirements,' British Standards Institution, 2002), (BS EN 13706–3:2002. 'Reinforced plastics composites. Specifications for pultruded profiles. - Part 3: Specific requirements,' British Standards Institution, 2002), (TR57 Strengthening Concrete Structures With Fibre Composite Materials: Acceptance, Inspection And Monitoring, The Concrete Society, UK., 2003)
	Denmark	(Fiberline Design Manual, 2003)
	Finland	(Clarke, 1996)
	France	(Clarke, 1996)
	Sweden	(Clarke, 1996)
	Netherlands	(CUR 96. Fiber Reinforced Polymers in Civil Load Bearing Structures, 2003)
	Italy	(Grimaldi, 2007), (Boscato <i>et al.</i> , 2017)
	USA	(American Society for Civil Engineers, 2010), (BS EN 13706–1:2002. 'Reinforced plastic composites - Specification for pultruded profiles - Part 1: Designation,' British Standards Institution, 2002), (AC 125 Acceptance Criteria for Concrete and Reinforced and Unreinforced Masonry Strengthening Using Fiber-Reinforced Polymer (FRP) Composite Systems, 2001), (AC 125 ACCEPTANCE CRITERIA FOR CONCRETE AND REINFORCED AND UNREINFORCED MASONRY STRENGTHENING USING EXTERNALLY BONDED FIBER-REINFORCED POLYMER (FRP) COMPOSITE SYSTEMS, 2010), (AC 187 (2001)), (ACI 440.2R-02, 2002), (Fouad <i>et al.</i> , 2003), (Code of standard practice for fabrication and installation of pultruded FRP structures,' ANSI standard, American Composites Manufacturers Association, 1st Edition, Arlington, VA. 2012., 2012), (Structural design of FRP components,' CTI Bulletin ESG-152 (13), Cooling Technology Institute, 2013), (Fibreglass pultruded structural products for use in cooling towers,' CTI Code Tower - Standard Specification, CTI Bulletin STD-137, Cooling Technology Institute, 2013, no date), (Standard definitions of terms relating to reinforced plastic pultruded products, D3918–96 (2003), ASTM, 2003), (Standard test method for shear properties of composite materials by the –notched beam method, D5379–05, ASTM, 2005), (ASTM International, 2013), (Standard test method for compressive residual strength properties of damaged polymer polymer matrix composite plates,' D7290–06, ASTM, 2006), (Standard test method for indentation hardness of rigid plastics by means of a barcol impressor,' D2583, ASTM, 2007), (Standard test method for deflection temperature of plastics under flexural load,' D648–07, ASTM, 2007), (Standard test method for tension-tension fatigue of oriented fiber, resin matrix composites, ASTM D3479/D3479M - 96(2007), ASTM, 2007), (Standard test method for in-plane shear properties of composite laminates, D4255/D4255M - 01(2007), ASTM, 2007), (Standard test method for open-hole tensile strength of polymer matrix composite laminates,' D5766–07, ASTM, 2007), (ASTM, 2007), (Standard test method for compressive properties of rigid plastics,' D695–08, ASTM, 2008), (Standard test method for tensile properties of plastics,' D638–08, ASTM, 2008), (Standard test method for density and specific gravity (relative density) of plastics by displacement,' D792–08, ASTM, 2008), (Standard test method for ignition loss of cured reinforced resins,' D2584–08, ASTM, 2008), (Standard test method for tensile properties of polymer matrix materials,' D3039/ D3039M - 08, ASTM, 2008), (Standard test methods for compressive properties of unidirectional or crossply fiber-resin composites, D3410/D3410M-03(2008), ASTM, 2008), (Standard test method for transition temperature and enthalpies of fusion and crystallization of polymers by differential scanning calorimetry,' D3418–08, ASTM, 2008), (Standard specification for dimensional tolerance of thermosetting glass-reinforced plastic pultruded shape,' D3917–8, ASTM, 2008), (Standard practice for classifying visual defects in thermosetting plastic pultruded shapes,' D4385–08, ASTM, 2008), (Standard guide for testing polymer matrix composite materials,' D4762–08, ASTM, 2008), (Standard test method for bearing strength,' D953–09, ASTM, 2009), (Standard test method for tensile, compressive, and flexural creep and creep rupture of plastics,' D2990–09, ASTM, 2009), (Standard test method for determining the compressive properties of polymer matrix composite laminates using a combined loading compression (CLC) test fixture,' D6641/ D6641M-09, ASTM, 2009), (Standard test method for void content of reinforced plastics,' D2734–09, ASTM, 2009), (Standard practice for classifying reinforced plastic pultruded

Table 3 Continued

Approaches	References	
	shapes according to composition,' D3647–09, ASTM, 2009), (Standard practice for testing pultruded composites,' D7745–11, ASTM, 2011), (Bedford Reinforced Plastics Design Guide, 2012), (Creative Pultrusions, 2017), (Strongwell design manual, 2010)	
Germany	(DIN EN 13121. <i>Structural Polymer Components for Building and Construction</i> , 2010), (BÜV-Tragende Kunststoff Bauteile im Bauwesen TKB – Richtlinie für Entwurf, Bemessung und Konstruktion, 2019)	
Japan	(Japan Society of Civil Engineers (JSCE), 2001), (JSCE Recommendation for Upgrading of Concrete Structures with use of Continuous Fiber Sheets, Concrete Engineering Series 41, Japan Society of Civil Engineers, 2001)	
Canada	(CSA Specification for Fiber-Reinforced Polymers, CSA-S807–10, Canadian Standards Association (CSA) International, 2010), (ISIS Design Manual No. 4 – FRP Rehabilitation of Reinforced Concrete Structures, ISIS Canada., 2001), (ISIS Durability Monograph – Durability of Fiber Reinforced Polymers in Civil Infrastructure, ISIS Canada., no date)	
Switzerland	(FIB Externally Bonded FRP Reinforcement for RC Structures, International Federation for Structural Concrete, 2001)	
Belgium	(CEN Reinforced Plastic Composites: Specifications for Pultruded Profiles, Parts 1–3, EN 13706, 2002)	
Brazil	(Petroleum and natural gas industries - pultruded shapes - Part 1: materials, test methods and dimensional tolerances,' Associacao Brasileira de Normas Tecnicas – ABNT, NBR 15708–1: 2011, Rio de Janeiro, Brazil, 2011), (Petroleum and natural gas industries - pultruded shapes - Part 5: structural shapes,' Associacao Brasileira de Normas Tecnicas – ABNT, NBR 15708–5: 2011, Rio de Janeiro, Brazil, 2011)	
Year of publication	Before 2000	(Clarke, 1996), (BS EN 13706–1:2002. 'Reinforced plastic composites - Specification for pultruded profiles - Part 1: Designation,' British Standards Institution., 2002)
	2000–2009	(Grimaldi, 2007), (The Highways Agency, 2005), (TR 55 Design Guidance for Strengthening Concrete Structures Using Fiber Composite Materials, 2004), (TR57 Strengthening Concrete Structures with Fiber Composite Materials: Acceptance, Inspection and Monitoring, 2003), (BS EN 13706–1:2002. 'Reinforced plastic composites - Specification for pultruded profiles - Part 1: Designation,' British Standards Institution., 2002), (BS EN 13706–2:2002. 'Reinforced plastic composites - Specification for pultruded profiles - Part 2: Methods of test and general requirements,' British Standards Institution, 2002), (TR57 Strengthening Concrete Structures WithFibre Composite Materials: Acceptance, Inspection And Monitoring, The Concrete Society, UK., 2003), (Fiberline Design Manual, 2003), (CUR 96. Fiber Reinforced Polymers in Civil Load Bearing Structures, 2003), (AC 125 Acceptance Criteria for Concrete and Reinforced and Unreinforced Masonry Strengthening Using Fiber-Reinforced Polymer (FRP) Composite Systems, 2001), (AC 187 (2001)), (ACI 440.2R-02, 2002), (Fouad <i>et al.</i> , 2003), (Standard definitions of terms relating to reinforced plastic pultruded products, D3918–96(2003), ASTM, 2003), (Standard test method for shear properties of composite materials by the –notched beam method, D5379–05, ASTM, 2005), (Standard test method for compressive residual strength properties of damaged polymer polymer matrix composite plates,' D7290–06, ASTM, 2006), (Standard test method for indentation hardness of rigid plastics by means of a barcol impressor,' D2583, ASTM, 2007), (Standard test method for in-plane shear properties of composite laminates, D4255/D4255M - 01(2007), ASTM, 2007), (Standard test method for open-hole tensile strength of polymer matrix composite laminates,' D5766–07, ASTM, 2007), (ASTM, 2007), (Standard test method for compressive properties of rigid plastics,' D695–08, ASTM, 2008), (Standard test method for tensile properties of plastics,' D638–08, ASTM, 2008), (Standard test method for density and specific gravity (relative density) of plastics by displacement,' D792–08, ASTM, 2008), (Standard test method for ignition loss of cured reinforced resins,' D2584–08, ASTM, 2008), (Standard test method for tensile properties of polymer matrix materials,' D3039/D3039M - 08, ASTM, 2008), (Standard test methods for compressive properties of unidirectional or crossply fiber-resin composites, D3410/D3410M-03(2008), ASTM, 2008), (Standard test method for transition temperature and enthalpies of fusion and crystallization of polymers by differential scanning calorimetry,' D3418–08, ASTM, 2008), (Standard specification for dimensional tolerance of thermosetting glass-reinforced plastic pultruded shape,' D3917–8, ASTM, 2008), (Standard practice for classifying visual defects in thermosetting plastic pultruded shapes,' D4385–08, ASTM, 2008), (Standard guide for testing polymer matrix composite materials,' D4762–08, ASTM, 2008), (Standard test method for bearing strength,' D953–09, ASTM, 2009), (Standard test method for tensile, compressive, and flexural creep and creep rupture of plastics,' D2990–09, ASTM, 2009),
	(Continued)	

(Continued)

Table 3 Continued

Approaches	References
2010–2015	(Standard test method for determining the compressive properties of polymer matrix composite laminates using a combined loading compression (CLC) test fixture,' D6641/D6641M-09, ASTM, 2009), (Standard test method for void content of reinforced plastics,' D2734-09, ASTM, 2009), (Standard practice for classifying reinforced plastic pultruded shapes according to composition,' D3647-09, ASTM, 2009), (Japan Society of Civil Engineers (JSCE), 2001), (JSCE Recommendation for Upgrading of Concrete Structures with use of Continuous Fiber Sheets, Concrete Engineering Series 41, Japan Society of Civil Engineers, 2001), (ISIS Design Manual No. 4 – FRP Rehabilitation of Reinforced Concrete Structures, ISIS Canada., 2001), (<i>FIB Externally Bonded FRP Reinforcement for RC Structures</i>, International Federation for Structural Concrete, 2001), (CEN Reinforced Plastic Composites: Specifications for Pultruded Profiles, Parts 1–3, EN 13706, 2002), (MBrace Composite Strengthening System: Engineering Design Guidelines, Master Builders, OH., 2006), (Bank <i>et al.</i>, 2003) (American Society for Civil Engineers, 2010), (AC 125 ACCEPTANCE CRITERIA FOR CONCRETE AND REINFORCED AND UNREINFORCED MASONRY STRENGTHENING USING EXTERNALLY BONDED FIBER-REINFORCED POLYMER (FRP) COMPOSITE SYSTEMS, 2010), (Code of standard practice for fabrication and installation of pultruded FRP structures,' ANSI standard, American Composites Manufacturers Association, 1st Edition, Arlington, VA. 2012., 2012), (Structural design of FRP components,' CTI Bulletin ESG-152 (13), Cooling Technology Institute, 2013), (Fibreglass pultruded structural products for use in cooling towers,' CTI Code Tower - Standard Specification, CTI Bulletin STD-137, Cooling Technology Institute, 2013, 2017), (ASTM International, 2013), (Standard test method for tension-tension fatigue of oriented fiber, resin matrix composites, ASTM D3479/D3479M - 96(2007), ASTM, 2007), (Standard practice for testing pultruded composites,' D7745-11, ASTM, 2011), (Bedford Reinforced Plastics Design Guide, 2012), (Strongwell design manual, 2010), (<i>DIN EN 13121. Structural Polymer Components for Building and Construction</i>, 2010), (BÜV-Tragende Kunststoff Bauteile im Bauwesen TKB – Richtlinie für Entwurf, Bemessung und Konstruktion, 2019), (CSA Specification for Fiber-Reinforced Polymers, CSA-S807-10, Canadian Standards Association (CSA) International, 2010), (Petroleum and natural gas industries - pultruded shapes - Part 1: materials, test methods and dimensional tolerances,' Associacao Brasileira de Normas Tecnicas – ABNT, NBR 15708-1: 2011, Rio de Janeiro, Brazil, 2011), (Petroleum and natural gas industries - pultruded shapes - Part 5: structural shapes,' Associacao Brasileira de Normas Tecnicas – ABNT, NBR 15708-5: 2011, Rio de Janeiro, Brazil, 2011),
Area 2016–present Structural design and structural profiles in civil infrastructure	(CIRIA c779, 2018), (Creative Pultrusions, 2017) (Clarke, 1996), (Grimaldi, 2007), (American Society for Civil Engineers, 2010), (BS EN 13706-1:2002. 'Reinforced plastic composites - Specification for pultruded profiles - Part 1: Designation,' British Standards Institution., 2002), (BS EN 13706-2:2002. 'Reinforced plastic composites - Specification for pultruded profiles - Part 2: Methods of test and general requirements,' British Standards Institution., 2002), (TR57 Strengthening Concrete Structures With Fibre Composite Materials: Acceptance, Inspection And Monitoring, The Concrete Society, UK., 2003), (CUR 96. Fiber Reinforced Polymers in Civil Load Bearing Structures, 2003), (Code of standard practice for fabrication and installation of pultruded FRP structures,' ANSI standard, American Composites Manufacturers Association, 1st Edition, Arlington, VA. 2012., 2012), (Standard definitions of terms relating to reinforced plastic pultruded products, D3918-96(2003), ASTM, 2003), (Standard specification for dimensional tolerance of thermosetting glass-reinforced plastic pultruded shape,' D3917-8, ASTM, 2008), (Standard practice for classifying visual defects in thermosetting plastic pultruded shapes,' D4385-08, ASTM, 2008), (Standard practice for testing pultruded composites,' D7745-11, ASTM, 2011), (<i>DIN EN 13121. Structural Polymer Components for Building and Construction</i>, 2010), (BÜV-Tragende Kunststoff Bauteile im Bauwesen TKB – Richtlinie für Entwurf, Bemessung und Konstruktion, 2019), (CEN Reinforced Plastic Composites: Specifications for Pultruded Profiles, Parts 1–3, EN 13706, 2002), (CSA Specification for Fiber-Reinforced Polymers, CSA-S807-10, Canadian Standards Association (CSA) International, 2010), (ISIS Durability Monograph – Durability of Fiber Reinforced Polymers in Civil Infrastructure, ISIS Canada, no date), (Bank <i>et al.</i>, 2003)
Bridges and highway structures Design of structures in seismic zones	(CIRIA C779, 2018), (The Highways Agency, 2005), (Fouad, <i>et al.</i>, 2003) (Boscato <i>et al.</i>, 2017)

Table 3 Continued

Approaches	References
Strengthening systems and rehabilitation of concrete structures	(Arya <i>et al.</i> , 2004), (TR57 Strengthening Concrete Structures with Fiber Composite Materials: Acceptance, Inspection and Monitoring, 2003), (AC 125 Acceptance Criteria for Concrete and Reinforced and Unreinforced Masonry Strengthening Using Fiber-Reinforced Polymer (FRP) Composite Systems, 2001), (AC 125 ACCEPTANCE CRITERIA FOR CONCRETE AND REINFORCED AND UNREINFORCED MASONRY STRENGTHENING USING EXTERNALLY BONDED FIBER-REINFORCED POLYMER (FRP) COMPOSITE SYSTEMS, 2010), (AC 187 (2001)), (ACI 440.2R-02, 2002), (Japan Society of Civil Engineers (JSCE), 2001), (JSCE Recommendation for Upgrading of Concrete Structures with use of Continuous Fiber Sheets, Concrete Engineering Series 41, Japan Society of Civil Engineers, 2001), (ISIS Design Manual No. 4 – FRP Rehabilitation of Reinforced Concrete Structures, ISIS Canada., 2001), (FIB Externally Bonded FRP Reinforcement for RC Structures, International Federation for Structural Concrete, 2001)
Petroleum and natural gas industries – pultruded shapes	(Petroleum and natural gas industries - pultruded shapes - Part 1: materials, test methods and dimensional tolerances,' Associacao Brasileira de Normas Tecnicas – ABNT, NBR 15708–1: 2011, Rio de Janeiro, Brazil, 2011), (Petroleum and natural gas industries - pultruded shapes - Part 5: structural shapes,' Associacao Brasileira de Normas Tecnicas – ABNT, NBR 15708–5: 2011, Rio de Janeiro, Brazil, 2011)
Cooling towers	(Structural design of FRP components,' CTI Bulletin ESG-152 (13), Cooling Technology Institute, 2013), (Fibreglass pultruded structural products for use in cooling towers,' CTI Code Tower - Standard Specification, CTI Bulletin STD-137, Cooling Technology Institute, 2013, 2017)
Test methods	(Standard test method for shear properties of composite materials by the –notched beam method, D5379–05, ASTM, 2005), (ASTM International, 2013), (Standard test method for compressive residual strength properties of damaged polymer polymer matrix composite plates,' D7290–06, ASTM, 2006), (Standard test method for indentation hardness of rigid plastics by means of a barcol impressor,' D2583, ASTM, 2007), (Standard test method for deflection temperature of plastics under flexural load,' D648–07, ASTM, 2007), (Standard test method for tension-tension fatigue of oriented fiber, resin matrix composites, ASTM D3479/D3479M - 96(2007),ASTM, 2007), (Standard test method for in-plane shear properties of composite laminates,' D4255/D4255M - 01(2007), ASTM, 2007), (Standard test method for open-hole tensile strength of polymer matrix composite laminates,' D5766–07, ASTM, 2007), (ASTM, 2007), (Standard test method for compressive properties of rigid plastics,' D695–08, ASTM, 2008), (Standard test method for tensile properties of plastics,' D638–08, ASTM, 2008), (Standard test method for density and specific gravity (relative density) of plastics by displacement,' D792–08, ASTM, 2008), (Standard test method for ignition loss of cured reinforced resins,' D2584–08, ASTM, 2008), (Standard test method for tensile properties of polymer matrix materials,' D3039/D3039M - 08, ASTM, 2008), (Standard test methods for compressive properties of unidirectional or crossply fiber-resin composites,' D3410/D3410M-03(2008), ASTM, 2008), (Standard test method for transition temperature and enthalpies of fusion and crystallization of polymers by differential scanning calorimetry,' D3418–08, ASTM, 2008), (Standard guide for testing polymer matrix composite materials,' D4762–08, ASTM, 2008), (Standard test method for bearing strength,' D953–09, ASTM, 2009), (Standard test method for tensile, compressive, and flexural creep and creep rupture of plastics,' D2990–09, ASTM, 2009), (Standard test method for determining the compressive properties of polymer matrix composite laminates using a combined loading compression (CLC) test fixture,' D6641/D6641M-09, ASTM, 2009), (Standard test method for void content of reinforced plastics,' D2734–09, ASTM, 2009), (Standard practice for classifying reinforced plastic pultruded shapes according to composition,' D3647–09, ASTM, 2009)
Manufacturers' design manuals	(Fiberline Design Manual, 2003), (Bedford Reinforced Plastics Design Guide, 2012), (Creative Pultrusions, 2017), (Strongwell design manual, 2010)

or just on the one side in relation to the web (Bank, 2007):

$$A_{eff} = (t_w + t_f + t_{bp})L_{eff}. \quad (18)$$

The effective bearing length (L_{eff}) is accounted in the longitudinal direction of the pultruded element. It should be calculated considering either the width of the support or the length of the area where load is concentrated (Bank, 2007).

It is worth to notice that the mechanism accounts only for a crushing failure and not for local buckling of the web, which can happen due to the presence of concentrated load. More detailed procedure of the calculation accounting for both phenomena can be found in (Bank, 2007).

Though much effort was made to study transverse crushing of the web in metal structures, there are still a lot of missing points that need to be addressed when dealing with pultruded composites (Fernandes *et al.*, 2015a,b).

Axial loading: Tension and compression

The remarkable advantage of pultruded FRP elements is their high axial tensile strength as a result of the unidirectional position of the fibers within the profile. Tension members are those structural elements that are subjected to direct axial stress without significant flexure. During the service-life of the structure, stress concentrations due to discontinuities and reductions in the cross-sectional area can arise. This must be considered at the design stage of tension members providing sufficient resistance. As glass fiber composites have a relatively low Young's modulus, the axial strain can be significant. In the case of an axial tensile load imposed on the pultruded structural element, design value of axial tensile load ($N_{t,Sd}$) should be not more than the design value of the axial tensile resistance ($N_{t,Rd}$) (Grimaldi, 2007):

$$N_{t,Sd} \leq N_{t,Rd}. \quad (19)$$

In the case of the axial compressive load imposed on the pultruded structural element, the design value ($N_{c,Sd}$) should be not more than the design value of the axial tensile resistance ($N_{c,Rd}$) (Grimaldi, 2007):

$$N_{c,Sd} \leq N_{c,Rd}, \quad (20)$$

where $N_{c,Rd}$ can be obtained from (Grimaldi, 2007):

$$N_{c,Rd} = \min\{N_{c,Rd1}, N_{c,Rd2}\}. \quad (21)$$

$N_{c,Rd1}$ is the value of the compressive force acting on the cross-section of the pultruded element. It can be estimated as follows (Grimaldi, 2007):

$$N_{c,Rd1} = A \cdot \sigma_{c,d}, \quad (22)$$

where $\sigma_{c,d}$ is the design compressive strength of the material, A represents the area of the cross-section of the profile and $N_{c,Rd2}$ is the design compression value of the forces initiating the instability of the element. It can be estimated by testing or numerical/analytical modeling.

Table 2 shows primary studies (experimental, analytical, and numerical) investigating the influence of the fiber/matrix type and cross-sectional characteristics on the tensile/compressive behavior of pultruded elements due to different load cases.

Design Guidance and Standards

Design codes make it possible to realize civil engineering structures with a high level of safety and confidence. Currently, the construction market is experiencing accelerated growth of FRP composite applications. The process of standard and guideline development has already been initiated in some countries. The 1980s marked the starting point of this process. This procedure has been launched by considering their differences in mechanical and physical properties compared to conventional materials. The performance uncertainty of FRP materials can be significantly limited by to the appearance of common-based testing procedures together with material identification schemes. A single set of guidelines can be developed, as sufficient theoretical and practical experience has been already gained, owing to the FRP-based projects realized all over the world. The development of standards and codes is an ongoing process and is expected to even accelerate in the coming years.

Sudden unexpected loading events such as car accidents, explosions, or even terroristic attacks can lead to the so-called progressive failure. Such possible situations when failure of one structure within the whole building is followed by a domino-effect collapse of other structures is not accounted for in the current design codes. However, this is not a problem exclusive to composite structures; it also exists for concrete and steel ones (Stylianidis and Nethercot, 2017). This topic still remains poorly investigated. Therefore, various relevant indicators, coefficients, and equations must be included in the developing composite design codes to deal with such type of structural occasions. Specific subarticles within the guidelines are to be established in order to minimize or even avoid enormous damages resulting from progressive failure (Abdelwahed, 2019), (Jiang and Li, 2018).

Table 3 presents primary design guidance and standards of the pultruded structural profiles, listing the countries where they were developed, years of publication, and areas of applications.

Conclusions and Future Trends

Residual stresses within the structure can be easily evaluated with the help of fiber optic sensor technology, especially with the cost of sensors drastically decreased in the last decades. Although this technique has already been implemented for inspection and monitoring of FRP bars, it would be of great interest in the coming future to install actuators allowing a distant control of the composite elements (Correia, 2013).

Peculiarities of pultrusion process and the nature of composites impose certain limitations on the quality of the final product, namely, process induced shape distortions and residual stresses. Therefore, part of the elements produced do not pass quality control, while another part requires extra shimming operations during the assembly process. These additional manipulations result in decreased mechanical performance of pultruded structures (Abouhamzeh *et al.*, 2015). For a better optimized design,

appropriate consideration of these geometrical deviations is required (Kappel, 2018). Experimental as well as numerical studies regarding the mentioned problems are currently underway. Authors of this review paper intend to address some of these problems in their future studies. However, there are still a lot of missing points at the moment (Ding *et al.*, 2019). Studies relating process parameters and composite architecture with the resultant process induced shape distortions are still necessary.

In the case of local buckling of pultruded I-profiles, contemporary analytical methods represent their web and flanges as orthotropic plates separated from each other. Local buckling capacity of the profile is greatly affected by the fact that in the web-flange junction these plates affect each other by restraining rotational displacement to some extent. Development of more precise closed-form equations for the proper consideration of the mentioned effect is needed since all the existing methods are strictly restricted by certain approximations (Nunes, Silvestre and Correia, 2017), (Ragheb, 2017). It is of great interest to examine the case when this happens. Besides, the loss of stability can be viewed from a completely different perspective. Novel structural and mechanical systems can be designed where the buckling effect is considered as a positive source of energy for motion-related applications, and not as the detrimental effect (Hu and Burgueño, 2015).

Current technological level allows engineers to collect and analyze data related to the structural performance of the element at any specific point in the lifecycle (Skels *et al.*, 2018), (Tuloup *et al.*, 2019), (Amafabia *et al.*, 2017). This technique is called Structural Health Monitoring (SHM). It became a reality due to a wide range of available integrated devices such as resistance strain gauges, fiber optic sensors, piezoelectric sensors, eddy current sensors and micro-electromechanical systems sensors (Christof *et al.*, 2015), (Ramakrishnan *et al.*, 2016). This novel technique makes it possible to identify, locate, evaluate, and predict possible failures (Gomes *et al.*, 2018). Carbon-based composites obviate the need in extra sensors due to the nature of this material. Carbon composites form a conductive carbon material network (Roh *et al.*, 2016) that possesses the piezoresistivity effect. This will require further research in order to advance the state of the art in SHM further on (Giurgiutiu, 2015).

Initially, cross-sections of pultruded structural profiles were designed based on the corresponding sections of their metal counterparts. However, pultruded profiles exhibit completely different behavior, which makes the development and optimization of material-adapted shapes the matter of great importance. Consequently, the profile connection technologies should be developed as well. In the attempt of creating affordable, lightweight profiles with good thermal insulating properties, composite manufacturers are developing profiles with the core consisting of low-density and low-price materials (Correia, 2013).

Random variations of parameters within the processes (structural, chemical, or kinetic) related to material science due to random effects can be described by statistic term of stochasticity. Hull *et al.* highlighted six large categories of fluctuations: equilibrium (thermodynamic), structural/compositional, kinetic, frustration/degeneracy, imprecision in measurements, and uncertainties in modeling and simulation. Practically, speaking of materials in general and of composites in particular, these variations play a remarkable role. Material performance can be easily improved if manufacturers are able to understand, analyze, and handle this stochasticity (Hull *et al.*, 2018).

The main purpose of scientists involved in engineering tasks is to pursue the optimal working conditions to achieve optimal final products. In the recent years, the numerical optimization tools have been successfully employed to detect optimal working conditions in many different fiber reinforced polymers manufacturing processes (Struzziero *et al.*, 2019). In the case of pultrusion, the numerical optimization can be applied to the process parameters (Carlone *et al.*, 2007) as well as to the geometrical topology of the final product (Safonov, 2019). Multi-objective optimization of pultrusion still needs further investigations to fulfill the industrial requirements, therefore, it is a promising topic for future research.

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Assessment of the Added Value of Multiscale Modeling of Concrete for Structural Analysis of Segmental Tunnel Rings

Jiao-Long Zhang, Tongji University, Shanghai, China and TU Wien, Vienna, Austria

Eva Binder, Tongji University, Shanghai, China; TU Wien, Vienna, Austria; and Linnaeus University, Växjö, Sweden

Xian Liu and Yong Yuan, Tongji University, Shanghai, China

Herbert Mang, Tongji University, Shanghai, China and TU Wien, Vienna, Austria

Bernhard LA Pichler, TU Wien, Vienna, Austria

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Introduction

Concrete is a hierarchically organized material. Its material behavior is related to thermo-hygro-chemo-mechanical processes occurring at its microstructure (Zhang *et al.*, 2013). Thus, it is reasonable to predict its macroscopic properties by making use of information at smaller scales. In the framework of multiscale modeling of concrete, several homogenization methods have been developed, involving e.g., continuum micromechanics (Zaoui, 2002; Bernard *et al.*, 2003), asymptotic expansion techniques (Cui and Yang, 1996; Zhang *et al.*, 2015), lattice approach (Schlangen, 1993), and numerical homogenization by means of the finite element method (Geers *et al.*, 2010). Several homogenization schemes, based on continuum micromechanics, have been developed at the Institute for Mechanics of Materials and Structures of Vienna University of Technology. They allow for estimation of elastic properties (Hellmich and Mang, 2005), strength (Pichler and Hellmich, 2011; Königsberger *et al.*, 2018), creep (Scheiner and Hellmich, 2009; Königsberger *et al.*, 2016), shrink-age (Pichler *et al.*, 2007), and thermal expansion (Wang *et al.*, 2018b) of concrete. In order to assess the added value resulting from the use of such multiscale material models for concrete in the framework of structural analysis of reinforced concrete structures, the authors have been involved in a research project, entitled “Bridging the gap by means of multiscale structural analysis” (Mang, 2015). This contribution refers to a part of the results from this project.

A reliable assessment of the added value of multiscale structural analyses requires a comparison of experimental data with results from both conventional and multi-scale structural analysis, see Fig. 1. As for the present contribution, experimental results are taken from a real-scale test of a segmental tunnel ring (Fig. 2), carried out at Tongji University. Both conventional and multiscale structural analysis are based on transfer relations, representing analytical solutions of the linear theory of circular arches (Zhang *et al.*, 2018, 2019b, 2020). Concerning conventional structural analysis, the material properties of concrete are obtained from the formulae in the *fib* Model Code 2010 (Taerwe and Matthys, 2013). As regards multiscale structural analysis, micromechanics-based multiscale models are taken from Königsberger *et al.* (2018) and Hlobil (2016). This selection is inspired by the previous finding that the performance of these multiscale models for evaluation of early-age material properties of the concrete used for the Hong Kong-Zhuhai-Macao Bridge (HZMB) is significantly better than that based on the *fib* Model Code (Wang *et al.*, 2018a), see Fig. 3. This also provides the motivation to quantify the added value of these multiscale models in the framework of structural analysis.

The main content of the present contribution is as follows: (1) hierarchical organization of the analyzed segmental tunnel ring, (2) evaluation of the material properties of concrete by means of multiscale modeling, (3) estimation of the effective stiffness of elements with crack bands and of crack opening widths, (4) transfer relations representing analytical solutions of the linear theory of circular arches, (5) assessment of the added value of multiscale structural analysis, and (6) conclusions drawn from the present study.

Hierarchical Organization of the Segmental Tunnel Ring

Herein, by segmental tunnel ring, one of the rings tested at Tongji University (Liu *et al.*, 2016; Zhang *et al.*, 2019a) is meant, see Fig. 2. The radius of the axis of the ring, R , is equal to 2.925 m. The ring consists of six reinforced concrete segments. The six joints of these segments are positioned at angular coordinates $\varphi_j = i, 2, \dots, 6$ equal to $8^\circ, 73^\circ, 138^\circ, 222^\circ, 287^\circ$, and 352° , where φ denotes the angular coordinate, measured from the center of segment ①. The thickness H and the width B of the segments amount to 35 cm and 1.2 m, respectively.

Both the segments and the microstructure of concrete are hierarchically organized, see Fig. 4. At the structural level, the six segments are subdivided into 34 elements. Their tangential length is set equal to the crack spacing $\ell_{cs} \approx 50$ cm, obtained from experimental measurements. As regards the element level, each element, consisting of reinforcement and concrete, is either intact or containing one central crack band, flanked by lateral undamaged domains. The reinforcement drawing is contained in (Zhang *et al.*, 2019a). Young's modulus E_s and the yield stress f_y of the reinforcement were equal to 210 GPa and 335 MPa, respectively. The concrete was produced from cement, fly ash, slag, water, sand, and aggregates, see Table 1 for their dosages. The maximum size of the aggregates, d_{max} , was equal to 2 cm. The uniaxial compressive strength of concrete, f_c , reached 28 days after production, amounted to 58 MPa. Multiscale analysis of concrete involved six scales (Zhang *et al.*, 2019a). Scale I refers to C-S-H gel consisting of gel pores that are embedded in a matrix of solid C-S-H. Scale II is associated with the hydration foam, consisting of C-S-H gel needles and spherical capillary pores filled with water or air. The needles and the pores are randomly distributed in space. Scale III refers to the cement paste, consisting of a matrix of hydrate foam and spherical inclusions, representing unhydrated cement, slag,

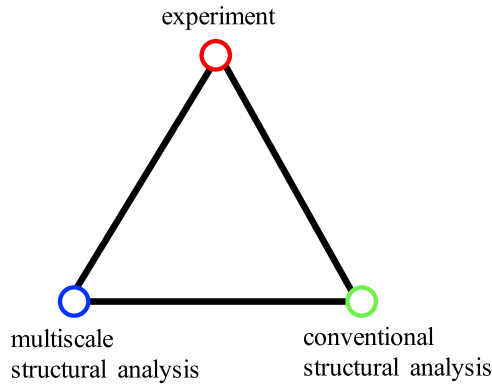


Fig. 1 Triad of results required for an assessment of the added value of multiscale analyses of tunnel segments. Reproduced from Mang, H., 2015. Bridging the gap by means of multiscale structural analyses, Research project supported by the Austrian Science Fund (FWF) P281 31-N32.

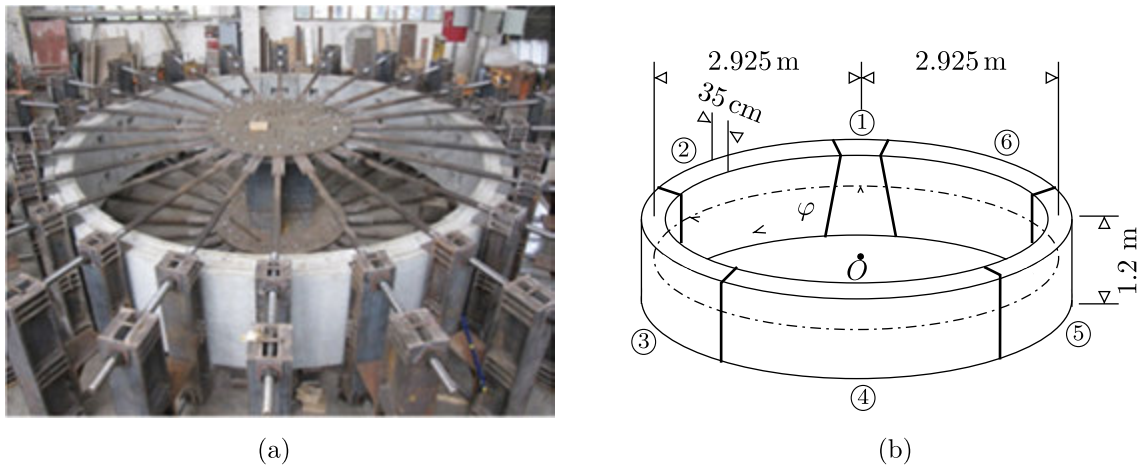


Fig. 2 (a) Photo and (b) geometric dimensions of the investigated segmental tunnel ring according to Liu *et al.* (2016) and Zhang *et al.* (2017), respectively. ①-⑥ are the numbers of the segments, and φ stands for the angular coordinate, measured from the center of the segment ①. According to Liu, X., Bai, Y., Yuan, Y., Mang, H., 2016. Experimental investigation of the ultimate bearing capacity of continuously jointed segmental tunnel linings. *Structure and Infrastructure Engineering* 12, 1364–1379. Zhang, J.L., Vida, C., Yuan, Y., *et al.*, 2017. A hybrid analysis method for displacement-monitored segmented circular tunnel rings. *Engineering Structures* 148, 839–856.

and fly ash. Scale IV is associated with the mortar, consisting of a matrix of cement paste and spherical inclusions of sand. Scale V refers to sane concrete, consisting of a matrix of mortar and spherical inclusions of aggregates. Scale VI refers to cracked concrete, containing a matrix of concrete and penny-shaped inclusions of parallel cracks.

Evaluation of the Material Properties of Concrete by Means of Multiscale Modeling

Conventionally, the material properties of concrete can be obtained e.g., from the formulae of the *fib* Model Code 2010 (Taerwe and Matthys, 2013), see details in the Appendix. Based on the experimentally determined compressive strength, $f_c = 58$ MPa, a stress-strain diagram for concrete in tension is obtained, see Fig. 5(a). Alternatively, the material properties of the concrete can also be evaluated by means of multiscale modeling, see Fig. 5(b) for the obtained stress-strain diagram. Details are described in the following.

Homogenization of the stiffness and strength of the concrete is carried out, using the model by Königsberger *et al.* (2018). It allows for consideration of the hierarchical organization of the material, the shape, the volume fractions, and the material constants of the heterogeneities, as well as for their mutual interaction at different scales of observation. The hierarchical organization of the material and the shape of the constituents are shown in Fig. 4. The corresponding volume fractions of the material constituents of concrete are quantified, based on the initial composition of the concrete, see Table 1, and the hydration degrees of cement, fly ash, and slag, see details in (Zhang *et al.*, 2019a). Herein, the hydration degrees ξ of the cement, the slag, and the fly ash are estimated as $\xi_{\text{cement}} = 0.8$ (Termkhajornkit *et al.*, 2014), $\xi_{\text{slag}} = 0.4$ (Lumley *et al.*, 1996), $\xi_{\text{flyash}} = 0$, respectively, considering that the real-scale test was carried out 28 days after casting of the segments. The mutual interactions of the material constituents are considered in the homogenization schemes. In more detail, matrix-inclusion composites, such as at Scales I, III, IV, and V, are

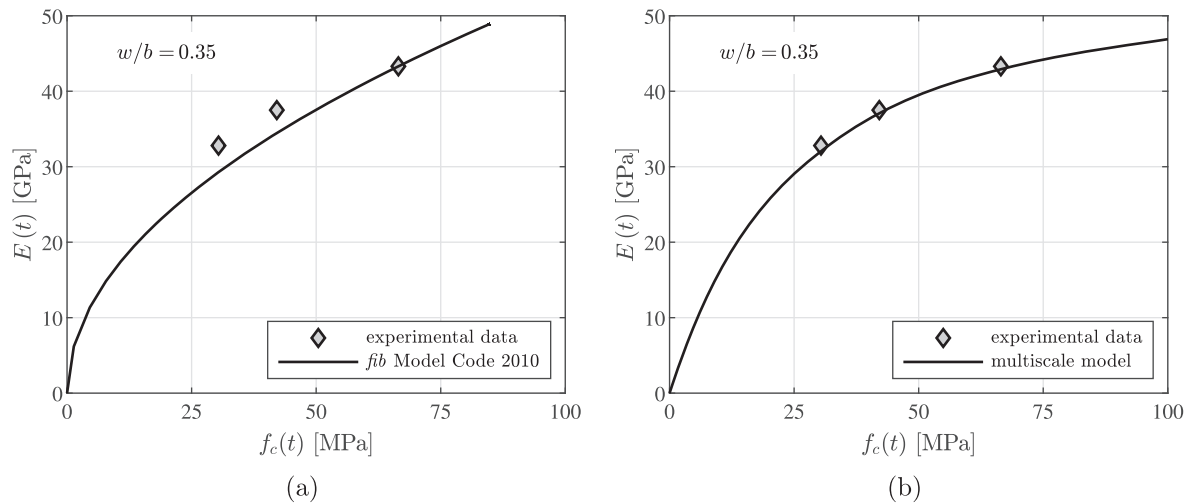


Fig. 3 Correlation between the elasticity modulus E and the compressive strength f_c of the concrete used for the immersed tunnel connecting the two parts of the Hong Kong-Zhuhai-Macao Bridge: (a) comparison of the results from the *fib* Model Code (Taerwe and Matthys, 2013) with the experimental results and (b) comparison of the results from multiscale analysis with the experimental results. Reproduced from Wang, H., Binder, E., Mang, H., Yuan, Y., Pichler, B., 2018a. Multiscale structural analysis inspired by exceptional load cases concerning the immersed tunnel of the Hong Kong-Zhuhai-Macao Bridge. *Underground Space* 3, 252–267.

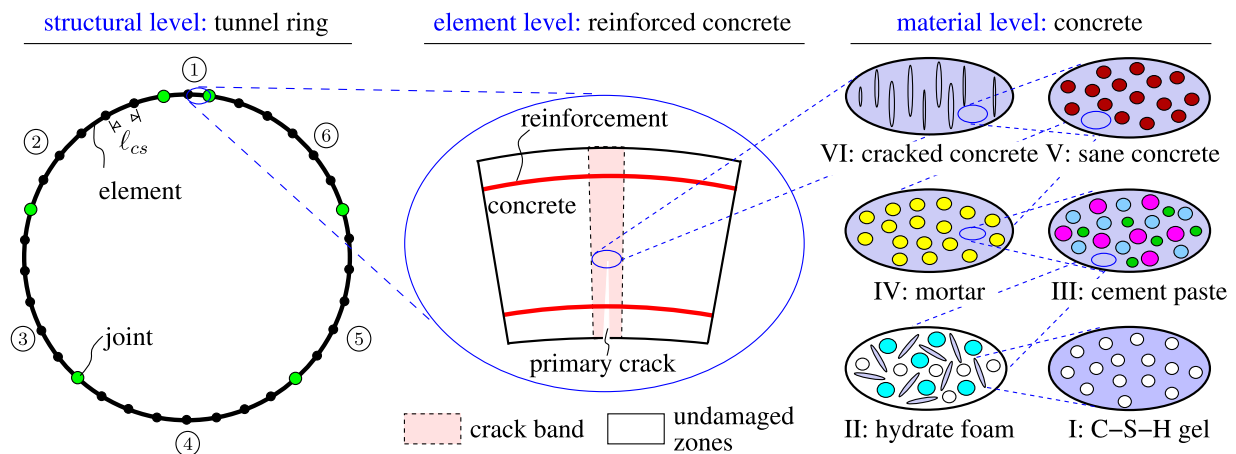


Fig. 4 Multiscale organogram of the segmental tunnel ring, tested at Tongji University. Reproduced from Zhang, J.L., Mang, H.A., Liu, X., Yuan, Y., Pichler, B., 2019a. On a nonlinear hybrid method for multiscale analysis of a bearing-capacity test of a real-scale seg-mental tunnel ring. *International Journal for Numerical and Analytical Methods in Geomechanics* 43, 1343–1372.

Table 1 Composition of the concrete used for the production of the segments

	Cement	Fly ash	Slag	Water	Sand	Aggregates
Dosage [kg/m ³]	323	67	57	152	631	1169

Note: Zhang, J.L., Mang, H.A., Liu, X., Yuan, Y., Pichler, B., 2019a. On a nonlinear hybrid method for multiscale analysis of a bearing-capacity test of a real-scale seg-mental tunnel ring. *International Journal for Numerical and Analytical Methods in Geomechanics* 43, 1343–1372.

homogenized, using the Mori-Tanaka scheme (Mori and Tanaka 1973, Benveniste, 1987). The polycrystalline microstructure at Scale II is homogenized, using the self-consistent scheme (Hershey, 1954, Kröner, 1958). Homogenization of the elasticity modulus and the uniaxial compressive strength of concrete delivers the following results:

$$E = 43.6 \text{ GPa}, \quad (1)$$

$$f_c = 62.0 \text{ MPa}. \quad (2)$$

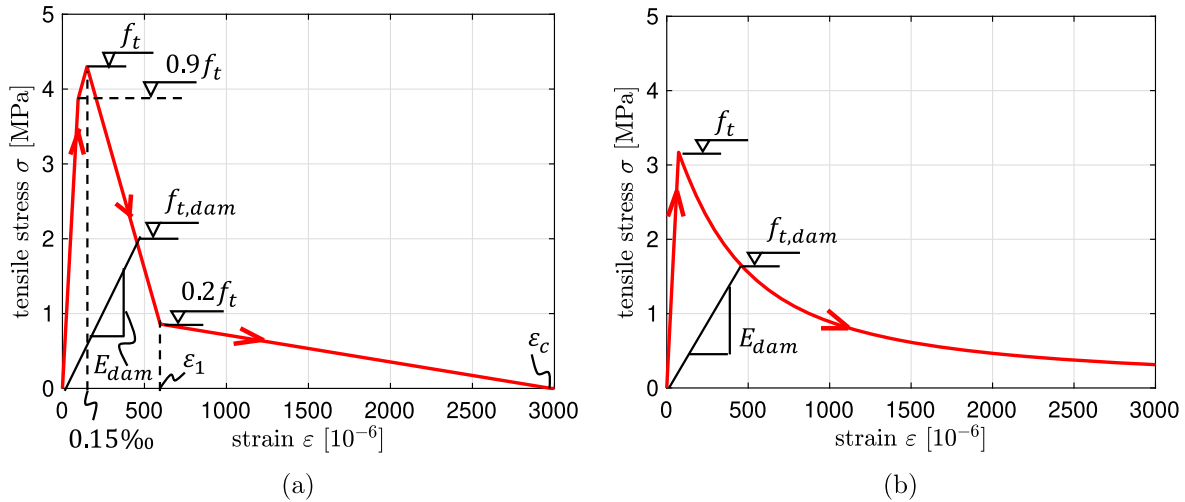


Fig. 5 Stress-strain diagrams for concrete in tension, computed (a) by the formulae of the *fib* Model Code (Taerwe and Matthys, 2013), where $\epsilon_1 = 0.000596$ and $\epsilon_c = 0.002980$ and (b) by the multiscale model by Hlobil (2016).

Notably, the values of the compressive strength of the concrete, obtained from the multiscale model and from the experiment, 62 MPa and 58 MPa, respectively, are close to each other. Thus the performance of the multiscale model by Königsberger *et al.* (2018) is satisfactory.

Homogenization of the uniaxial tensile strength and description of the progressive deterioration of the crack bands are based on the multiscale model by Hlobil (2016). It provides relations between the increase of the crack density ω and the decrease of the elastic stiffness E_{dam} and the tensile strength $f_{t,dam}$. The following results were obtained:

$$f_t = 3.17 \text{ MPa}, \quad (3)$$

$$E_{dam}(\omega) = \frac{1}{1 + 5.02\omega} \cdot E, \quad (4)$$

$$f_{t,dam}(\omega) = \sqrt{\frac{0.402}{0.402 + 0.396\omega}} \cdot f_t. \quad (5)$$

The combination of Eqs. (1) and (3)-(5) allows for determination of the stress-strain diagram for concrete in tension, see Fig. 5(b).

Estimation of Effective Stiffnesses of Elements and of Crack Openings

Cracks of the segments were observed after load step 4. Consideration of these cracks requires subdivision of the segment into "simulation elements", see Fig. 4. The extensional stiffness and the bending stiffness of the undamaged lateral zones of the elements are equal to their initial values, determination of which is based on the Bernoulli-Euler hypothesis and on linear-elastic behavior of the concrete and steel, both in compression and tension. As for the crack band, the extensional stiffness and the bending stiffness are decreasing with increasing damage. Determination of the state of damage requires an incremental-iterative solution procedure, see details in (Zhang *et al.*, 2019a). Evaluation of the extensional stiffness and the bending stiffness of the crack band is based on (i) the Bernoulli-Euler hypothesis, (ii) linear-elastic behavior of steel, both in tension and in compression, and (iii) linear-elastic behavior of concrete in compression as well as on linear-elastic, nonlinear-softening of concrete in tension, see also Fig. 5. The effective extensional stiffness, $\overline{EA}^{(e)}$, and bending stiffness, $\overline{EI}^{(e)}$, of the corresponding elements $e = 1, 2, \dots, 34$ are obtained by means of Voigt-Reuss-Hill estimates (Hill, 1952).

Crack opening at the surface of the elements, $w_c^{(e)}$, is estimated by multiplying the normal strain at the cracked surface of the corresponding crack band, $\epsilon_{suf}^{(e)}$, by the size of this zone, l_{cb} , i.e.,

$$w_c^{(e)} = l_{cb} \cdot \epsilon_{suf}^{(e)}. \quad (6)$$

Evaluation of $\epsilon_{suf}^{(e)}$ is based on the Bernoulli-Euler hypothesis. It implies that a linear distribution of the normal strain in the tangential direction prevails inside every crack band. Thus,

$$\epsilon_{suf}^{(e)} = \frac{\overline{N}^{(e)}}{\overline{EA}^{(e)} + \frac{\overline{M}^{(e)}}{\overline{EI}^{(e)}(\epsilon_{suf}^{(e)} - R)}}, \quad (7)$$

where $r_{\text{surf}}^{(e)} = [R + H/2; R - H/2]$ denotes the radial coordinate of the cracked surface, and $\bar{N}$ and $\bar{M}$ stand for the average axial force and the average bending moment over the simulation elements. The typical width of a crack band in the concrete, l_{cb} , suggested by Bažant and Oh (1983), amounts to three times the maximum aggregate size d_{max} , i.e.,

$$l_{cb} = 3d_{\text{max}}. \quad (8)$$

Therefore, the expression for the crack opening at the surface of the elements, $w_c^{(e)}$, is rewritten as

$$w_c^{(e)} = 3d_{\text{max}} \left[\frac{\bar{N}^{(e)}}{EA^{(e)} + \frac{\bar{M}^{(e)}}{EI^{(e)}(r_{\text{surf}}^{(e)} - R)}} \right]. \quad (9)$$

Transfer Relations as a Vehicle for Structural Analysis of Tunnel Rings

Segmental tunnel rings represent typical circular arch structures. Thus, the transfer relations, which are analytical solutions of the linear theory of circular arches, are used for structural analysis of such rings. These relations read as (Zhang *et al.*, 2017)

$$\begin{bmatrix} u(\varphi) \\ v(\varphi) \\ \theta(\varphi) \\ M(\varphi) \\ N(\varphi) \\ V(\varphi) \\ \hline 1 \end{bmatrix} = \begin{bmatrix} \cos \varphi & \sin \varphi & T_{13}(\varphi) & T_{14}(\varphi) & T_{15}(\varphi) & T_{16}(\varphi) & \sum u^L(\varphi) \\ -\sin \varphi & \cos \varphi & T_{23}(\varphi) & T_{24}(\varphi) & T_{25}(\varphi) & T_{26}(\varphi) & \sum v^L(\varphi) \\ 0 & 0 & 1 & T_{34}(\varphi) & T_{35}(\varphi) & T_{36}(\varphi) & \sum \theta^L(\varphi) \\ 0 & 0 & 0 & 1 & T_{45}(\varphi) & T_{46}(\varphi) & \sum M^L(\varphi) \\ 0 & 0 & 0 & 0 & \cos \varphi & -\sin \varphi & \sum N^L(\varphi) \\ 0 & 0 & 0 & 0 & \sin \varphi & \cos \varphi & \sum V^L(\varphi) \\ \hline 0 & 0 & 0 & 0 & 0 & 0 & 1 \end{bmatrix} \begin{bmatrix} u_i \\ v_i \\ \theta_i \\ M_i \\ N_i \\ V_i \\ \hline 1 \end{bmatrix} \quad (10)$$

where

$$\begin{aligned} T_{13}(\varphi) &= R \sin \varphi, T_{14}(\varphi) = \frac{R^2}{EI} (\cos \varphi - 1), \\ T_{15}(\varphi) &= \frac{R}{EA} \frac{1}{2} \varphi \sin \varphi + \frac{R^3}{EI} \left(\frac{1}{2} \varphi \sin \varphi + \cos \varphi - 1 \right), \\ T_{16}(\varphi) &= \frac{R}{EA} \left(\frac{1}{2} \varphi \cos \varphi - \frac{1}{2} \sin \varphi \right) + \frac{R^3}{EI} \left(\frac{1}{2} \varphi \cos \varphi - \frac{1}{2} \sin \varphi \right), \\ T_{23}(\varphi) &= R (\cos \varphi - 1), T_{24}(\varphi) = \frac{R^2}{EI} (\varphi - \sin \varphi), \\ T_{25}(\varphi) &= \frac{R}{EA} \left(\frac{1}{2} \varphi \cos \varphi + \frac{1}{2} \sin \varphi \right) + \frac{R^3}{EI} \left(\varphi - \frac{3}{2} \sin \varphi + \frac{1}{2} \varphi \cos \varphi \right), \\ T_{26}(\varphi) &= \frac{R}{EA} \left(-\frac{1}{2} \varphi \sin \varphi \right) + \frac{R^3}{EI} \left(1 - \cos \varphi - \frac{1}{2} \varphi \sin \varphi \right), \\ T_{34}(\varphi) &= -\frac{R}{EI} \varphi, T_{35}(\varphi) = \frac{R^2}{EI} (\sin \varphi - \varphi), T_{36}(\varphi) = \frac{R^2}{EI} (\cos \varphi - 1), \\ T_{45}(\varphi) &= R (1 - \cos \varphi), T_{46}(\varphi) = R \sin \varphi. \end{aligned} \quad (11)$$

The vector on the left-hand-side of Eq. (10) contains the kinematic and static variables, referring to the cross-section at an arbitrary value of the angular coordinate φ . These variables are the radial displacement u , the tangential displacement v , the cross-sectional rotation θ , the bending moment M , the axial force N , and the shear force V . The vector on the right-hand-side of Eq. (10) contains the kinematic and static variables, referring to the initial cross-section (index "i"), i.e. $\varphi = 0$. These six quantities are the integration constants. Three of them, representing kinematic quantities, may be set equal to zero, i.e., $u_i = v_i = \theta_i = 0$. This is admissible because they describe rigid body displacement of the ring (Zhang *et al.*, 2019a). The three remaining integration constants, representing static quantities, are determined from three continuity conditions of a closed ring. The seven-by-seven matrix on the right-hand-side of Eq. (10) is the transfer matrix. The top-left six-by-six submatrix of this matrix represents the solution for an unloaded part of the segmental tunnel ring (Zhang *et al.*, 2017). The top six elements of the last column of the

transfer matrix stand for the superposition of so-called “load integrals” (superscript “ L ”). The latter represent analytical solutions for specific types of generalized loads. Load integrals for dead load, interfacial discontinuities of kinematic variables, and for point loads are given in Zhang *et al.* (2017). Load integrals for a uniform temperature change, ground pressure, and for the overload on the ground surface are given in Zhang *et al.* (2018), (2019b) and Zhang *et al.* (2021), respectively.

In this contribution, the hybrid method, developed by Zhang *et al.* (2017), (2019a), is employed for re-analysis of the real-scale test of the segmental tunnel ring, performed by Liu *et al.* (2016). The test was carried out in a force-controlled fashion. Compressive loading was imposed by 24 hydraulic jacks, resulting in point loads acting on the outer surface of the ring. The tangential displacement discontinuities across both the inner and the outer gaps of the joints and the vertical and the horizontal convergences were experimentally monitored. The hybrid method requires as input for the analysis (1) the point loads, imposed by the hydraulic jacks, and (2) the relative rotation angles at the joints, estimated from the measured displacement discontinuities.

The load integrals for a radial point load P , imposed at the position φ_p , read as (Zhang *et al.*, 2017):

$$u^L(\varphi) = \frac{1}{2} \left(\frac{PR}{EA} + \frac{PR^3}{EI} \right) \left[\sin(\varphi - \varphi_p) - (\varphi - \varphi_p) \cos(\varphi - \varphi_p) \right] H(\varphi - \varphi_p), \quad (12)$$

$$\begin{aligned} v^L(\varphi) &= \frac{PR}{EA} \left[\frac{1}{2} (\varphi - \varphi_p) \sin(\varphi - \varphi_p) \right] H(\varphi - \varphi_p) \\ &+ \frac{PR^3}{EI} \left[\frac{1}{2} (\varphi - \varphi_p) \sin(\varphi - \varphi_p) + \cos(\varphi - \varphi_p) - 1 \right] H(\varphi - \varphi_p), \end{aligned} \quad (13)$$

$$\theta^L(\varphi) = \frac{PR^2}{EI} [1 - \cos(\varphi - \varphi_p)] H(\varphi - \varphi_p), \quad (14)$$

$$M^L(\varphi) = -RP \sin(\varphi - \varphi_p) H(\varphi - \varphi_p), \quad (15)$$

$$N^L(\varphi) = P \sin(\varphi - \varphi_p) H(\varphi - \varphi_p), \quad (16)$$

$$V^L(\varphi) = -P \cos(\varphi - \varphi_p) H(\varphi - \varphi_p), \quad (17)$$

where $H(\varphi - \varphi_j)$ stands for the Heaviside function. The load integrals for the relative rotation angle, $\Delta\theta_j$, at the joint located at $\varphi = \varphi_j$, read as (Zhang *et al.*, 2017)

$$u^L(\varphi) = -R\Delta\theta_j \sin(\varphi - \varphi_j) H(\varphi - \varphi_j), \quad (18)$$

$$v^L(\varphi) = R\Delta\theta_j [1 - \cos(\varphi - \varphi_j)] H(\varphi - \varphi_j), \quad (19)$$

$$\theta^L(\varphi) = \Delta\theta_j H(\varphi - \varphi_j), \quad (20)$$

$$N^L(\varphi) = V^L(\varphi) = M^L(\varphi) = 0. \quad (21)$$

Assessment of the Added Value of Multiscale Structural Analysis Concerning the Prediction of Cracking

The assessment of the added value of multiscale modeling of concrete for structural analysis of segmental tunnel rings is based on the comparison of experimental data with results from both conventional and multiscale structural analysis, see Fig. 1. The focus is on two types of quantities that are relevant to the serviceability and the durability of tunnel linings:

- (1) The convergences in the vertical and the horizontal direction,
- (2) The opening widths of bending-induced cracks of concrete.

The experimental data refer to the first ten load steps of the previously mentioned real-scale test of a segmental tunnel ring, performed by Liu *et al.* (2016). These steps are associated with the increase of the external loading of the tunnel ring up to the level that simulates the ground pressure acting on the extrados of tunnel linings in the expected practical service (Liu *et al.*, 2016).

Two modes of structural analysis were carried out. The first one is a conventional structural analysis based on the *fib* Model Code, see the Appendix for the relevant formulae. The second one is a multiscale structural analysis based on the multiscale models for concrete, presented in Section “Evaluation of the Material Properties of Concrete by Means of Multiscale Modeling”. Both modes of analysis were nonlinear hybrid computations. They are *hybrid*, insofar as experimental data regarding the external loading and displacement measurements at the joints are used as input. They are *nonlinear*, because bending-induced cracking of the reinforced concrete segments is accounted for. Despite this nonlinearity, structural analysis can be subdivided into two load cases (Zhang *et al.*, 2019a). Load case I is concerned with the computation of the deformation of the segments of the tunnel ring, resulting from external loading, while the relative rotations at the joints are set equal to zero. Load case II aims at determination of rigid body displacements of the segments of the tunnel ring, resulting from the relative rotations at the joints. After estimation of these rotations based on the experimentally measured displacement discontinuities at the joints, they are postprocessed such that symmetric rigid body displacements of the segments are obtained (Zhang *et al.*, 2019a). In the given case, the restriction to

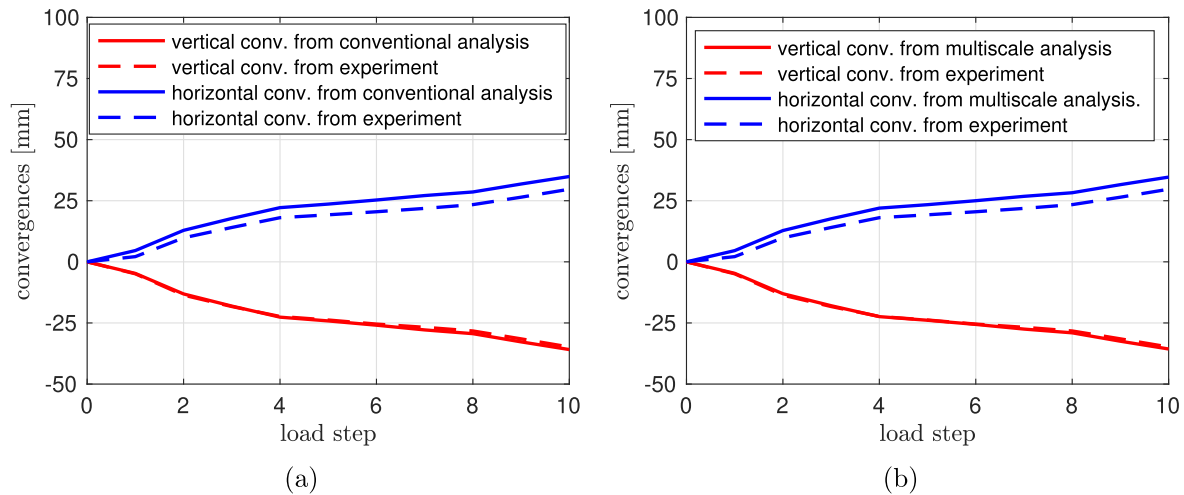


Fig. 6 Comparison of the convergences obtained from experimental measurements with those from (a) conventional structural analysis and (b) multiscale structural analysis.

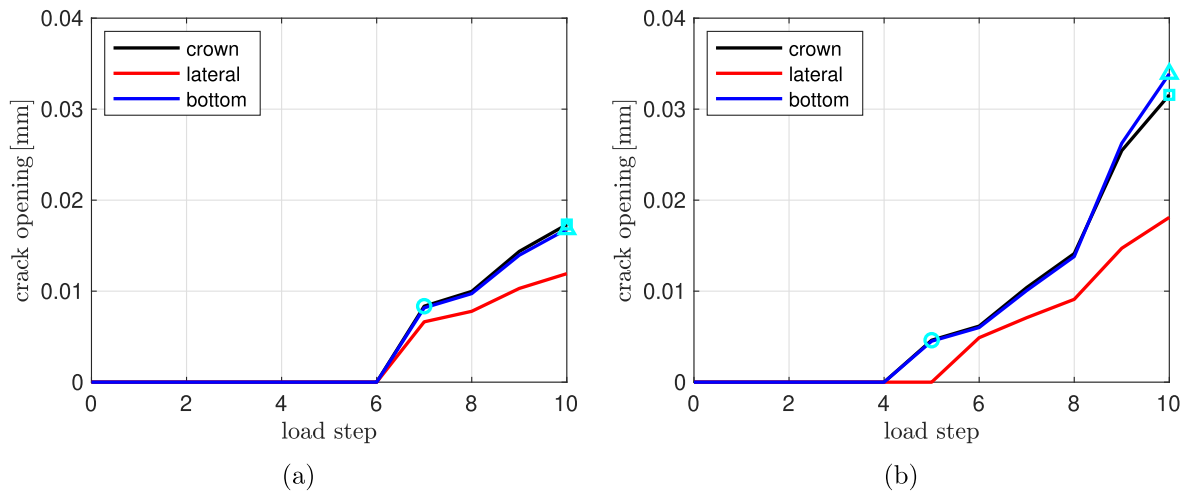


Fig. 7 Crack openings at the surface of the segments in the top element, the lateral element, and the bottom element, as a function of the load step: (a) conventional structural analysis and (b) multiscale structural analysis.

symmetric rigid body displacements is adequate, because the observed structural behavior was indeed virtually symmetric. This renders the available extension towards consideration of both symmetric *and* antisymmetric modes of rigid body displacements dispensable (Jiang *et al.*, 2021). Despite the nonlinearity of the material behavior in load case I, superposition of the results from the two load cases is admissible, because load case II only produces rigid body displacements and because the equilibrium equations are formulated for the undeformed configuration. This completes the nonlinear hybrid analysis.

As for the vertical and horizontal convergences, both conventional and multiscale structural analyses deliver very similar predictions which agree well with corresponding experimental measurements, see Fig. 6. Thus, both modes of structural analysis are useful. The added value resulting from the use of the multiscale model for concrete is insignificant. This was expected, because the convergences are governed by rigid body displacements resulting from the relative rotations at the joints, while the deformations of the segments is less important (Zhang *et al.*, 2019a).

As regards bending-induced cracks of concrete, the two modes of nonlinear hybrid analysis deliver different results. Conventional structural analysis indicates that cracking initiates at load step 7, see the cyan circle in Fig. 7(a). In the experiments, however, cracking was observed already at load step 5. Thus, conventional structural analysis overestimates the external loading at crack initiation by some 55%. Multiscale structural analysis, however, indicates that cracking starts already at load step 5, see the cyan circle in Fig. 7(b). This underlines the added value resulting from use of the multiscale model for concrete, as compared to the standard approach based on the *fib* Model Code. As for the final load step, i.e., load step 10, multiscale structural analysis results in crack opening widths at the crown and the bottom of the ring, which are approximately twice as large as the values obtained from conventional structural analysis, see the cyan squares and triangles in Fig. 7. Also the crack lengths are significantly different. In

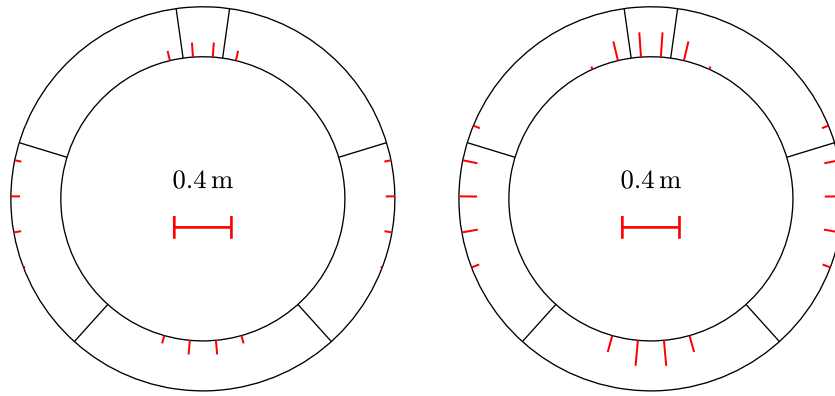


Fig. 8 Crack lengths at the final load step 10: obtained from (a) conventional structural analysis and (b) multiscale structural analysis.

case of conventional standard analysis, the maximum crack length is obtained in the region of the crown. It amounts to 10.2 cm, see Fig. 8(a). In case of multiscale analysis, the maximum crack length is obtained at the bottom of the ring. It amounts to 17.9 cm, see Fig. 8(b). It is concluded that multiscale structural analysis allows for a more realistic and more conservative durability assessment of segmental tunnel linings. The larger the crack opening widths and the larger the crack lengths, the higher is the risk that aggressive substances enter the segments and initiate corrosion of the steel reinforcement.

Thus, multiscale structural analysis is preferable for a durability-oriented design of segmental tunnel rings.

Conclusions

In order to assess the added value of multiscale structural analysis of segmental tunnel rings relative to a conventional mode of structural analysis, a real-scale test of a segmental tunnel ring, performed by Liu *et al.* (2016), was analyzed. The following conclusions are drawn from this analysis:

- (1) The input for the multiscale model for a customized concrete, used in this work, refers to its initial composition, defined by the mix design, and the maturity of the material, typically quantified by means of hydration degrees. Thus, efforts concerning material tests are superfluous. This is one of the merits of the multiscale model as compared to the *fib* Model Code.
- (2) Both conventional and multiscale structural analysis are equally reliable concerning estimation of the convergences. In that case, the added value of the multiscale structural analysis is insignificant, because the convergences are predominately governed by rigid body displacements of the segments, resulting from the relative rotation angles at the joints (Zhang *et al.*, 2019a), rather than by the deformation of the segments, which increases with bending-induced cracking of concrete.
- (3) Multiscale structural analysis resulted in correct predictions of the initiation of cracking of concrete. Results from conventional structural analysis, however, overestimated the level of the external loading at onset of cracking by some 55%. This underlines the added value of multiscale structural analysis. The difference between the two compared modes of structural analysis is significant for the long-term durability of segmental tunnel linings with a designed service life of 100 years and more.

Acknowledgments

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Appendix: Quantification of the Tensile Strength and of Softening of Concrete According to *fib* Model Code 2010 (Taerwe and Matthys, 2013)

The experimentally determined compressive strength of concrete, f_c , was equal to 58 MPa. Based on this value, the *fib* Model Code contains the following formulae for quantification of the tensile strength, f_t , the elasticity modulus, E , and the fracture energy, G , of concrete,

$$f_t = 2.12 \cdot \ln[1 + 0.1(f_c + 8)] = 4.30 \text{ MPa}, \quad (\text{A.1})$$

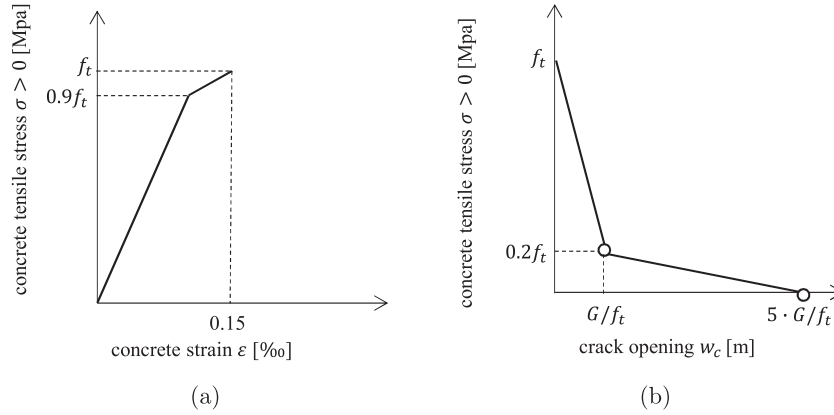


Fig. A.1 Schematic representation of the stress-strain and stress - crack opening relation for uniaxial tension, according to the *fib* Model Code 2010: (a) uncracked concrete and (b) cracked concrete. Reproduced from Taerwe, L., Matthys, S., 2013. *fib* Model Code for Concrete Structures 2010. Wiley (Ernst & Sohn).

$$E = 21.5 \cdot 1.0 \cdot \left(\frac{f_c + 8}{10} \right)^{1/3} = 40330 \text{ MPa}, \quad (\text{A.2})$$

$$G = 0.73 \cdot f_c^{0.18} = 154 \text{ N/m}. \quad (\text{A.3})$$

For uncracked concrete, the *fib* Model Code suggests a bilinear curve, see **Fig. A.1(a)**, describing the stress-strain relation. As for cracked concrete, another bilinear curve, see **Fig. A.1(b)**, describing the relation of the tensile stress and the crack opening, is given. Notably, the combination of Eqs. (6) and (8) allows for the evaluation of the tensile strain as a function of the crack opening. Considering Eqs. (A.1)–(A.3), (6), (8), $d_{\max} = 0.02 \text{ m}$, the stress-strain and the stress-crack opening relation, illustrated in **Fig. A.1**, results in a piecewise-linear curve, describing the stress-strain relation of both the uncracked and the cracked concrete in tension, see **Fig. 5(a)**.

The mathematical expressions for this stress-strain relation read as

$$\sigma = \begin{cases} E\varepsilon & \dots 0 < \varepsilon \leq \frac{0.9f_t}{E}, \\ 0.9f_t + \frac{0.1f_t}{0.00015 - 0.9f_t/E} \left(\varepsilon - \frac{0.9f_t}{E} \right) & \dots \frac{0.9f_t}{E} < \varepsilon \leq 0.00015, \\ f_t - \frac{0.8f_t}{\varepsilon_1 - 0.00015} (\varepsilon - 0.00015) & \dots 0.00015 < \varepsilon \leq \varepsilon_1, \\ 0.2f_t - \frac{0.2f_t}{\varepsilon_c - \varepsilon_1} (\varepsilon - \varepsilon_1) & \dots \varepsilon_1 < \varepsilon \leq \varepsilon_c, \\ 0 & \dots \varepsilon > \varepsilon_c, \end{cases} \quad (\text{A.4})$$

where $\varepsilon_1 = G/f_t = 0.000596$ and $\varepsilon_c = 5 \cdot G/f_t = 0.0029780$.

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Nature Based and Inspired Composite Materials: An Introduction

Mohamed El Mansori, Arts et Metiers Institute of Technology, Mechanics Surfaces and Materials Processing, HESAM Université, Châlons-en-Champagne, France and Texas A&M Engineering Experiment Station, Institute for Manufacturing Systems, College Station, TX, United States

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This article is an abridged introduction to the bio-composite section of the encyclopedia of material composites. The term “biocomposite” is used to denote fiber-reinforced polymer composite materials where the fibers and/or matrix are bio-based. Bio-composite materials derived from natural-renewable sources have received significant interest, mainly due to the increased awareness of and drive towards more environmentally sustainable technologies.

However, natural fiber quality is significantly influenced by harvesting and processing steps. There is a move to reduce the costs/steps of on-field processing to improve consistency in properties. The potential use of natural fiber composites (biocomposites) in industrial applications is directly related to the secondary manufacturing of these eco-friendly materials. Biocomposites, especially those with extended continuous fibers, present many challenges during near net shape processing where requirements include better dimensional tolerance, drilling holes for assembly, and controlled surface characteristics. At the outset, it is of primary importance to introduce both the elementary ideas concerning their multiscale structure, mechanical and fiber-matrix interface properties for fabricating parts from plant fibers to meet service requirements. Secondly, the cutting process of removing unwanted material in the production of biocomposite components is mandatory. However, to preserve their industrial functionalities (such as mechanical, thermal, acoustic, hygroscopic, or damping properties), these mechanical treatments face significant challenges. Research areas related to the observed performance to fundamentals of chemistry, material behavior, and the engineering sciences of heat transfer, solid mechanics and surface science (tribology) should be focused in detail.

The biocomposites section includes the development of the mechanics of natural fiber-based composites and their structural interpretations and chemical treatments. Cutting processes of bio-composites are complex primarily due to the nature of plant fibers. The basic two dimensional (orthogonal) cutting process is analysed in detail, followed by considerations of representative three-dimensional cutting operations such as drilling and milling. A simplified micromechanical model emphasizes various aspects of the cutting behavior of biocomposites such as thermal, material and surface considerations are operative simultaneously with varying degrees of importance depending on specific machining conditions.

The relative details with which bio-composites are processed reflects the current state of the art. Each article has been well focussed, and I hope that this will appeal to those who would not afford to read the biocomposites section as a whole in the order of presentation; it is aimed that the articles may prove to be helpful to researchers seeking an introduction to a part of the subject that is other than their speciality.

Finally, I am incredibly grateful to all the experts and colleagues; this section would not have been presented without their contributions.

Bio Composite Material: Review and its Applications in Various Fields

Piyush Patel, Piyush Gohil, and Vijay Parmar, The Maharaja Sayajirao University of Baroda, Baroda, Gujarat, India

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Introduction

A couple of decades back, request of buyers and enterprises for high performing materials and structures was so high for composite materials which drove it to the facing all around. As of late, the principal ecological problematical confronted today is because of the unbelievable generation and utilization of non-degradable plastics in each section of our lifecycle. Expanding worry about an unnatural weather change and exhausting oil holds have centered extraordinary piece of the logical examination to eco-composite materials that can be effectively debased. Accordingly, Researchers have turned out to be more intrigued in the advancement of ecologically agreeable biobased materials from renewable assets (Ashok Kumar *et al.*, 2017; Thakur *et al.*, 2012).

Bio based composite will be composite materials where plant *fibers* from timberland or agriculture are the fortifying constituent and *matrix* ought to ideally be bio based thermoplastic or thermoset. Bio-composites are the blend of regular strands, for example, wood filaments (hard wood and soft wood) or non-wood strands (hemp, sugarcane, rice straw, jute, banana, pine apple, oil palm, sisal, and flax) with polymer matrices from both of the sustainable and non-inexhaustible assets. Inexhaustible materials assets are presently picking up significance around the world (Bharath *et al.*, 2018; Mitra, 2014; Patel and Gohil, 2012; Netravali *et al.*, 2007).

Bio Composite Constituent

The term 'bio-composites' for the most part covers composite materials where in any event one segment ought to be bio-based;

- (1) Bio-polymers (e.g., PLA) reinforced by bio-fibers (jute)
- (2) Bio-fiber reinforced petroleum derived polymers which are non-biodegradable e.g., epoxy, polyolefin polyester, vinyl ester, phenolic.
- (3) Bio-polymers reinforced synthetic fibers such as glass or carbon (Zini and Scandola, 2011a; Mohanty *et al.*, 2001).

Biopolymers

These biodegradable polymers include renewable or synthetic input materials for production. Classification of natural bio-degradable polymers are based on the basis of sources; (1) Directly extracted from biomass. (for example, Starch, cellulose, casein etc.) (2) Classically synthesized from bio derived monomers (for example, Polylactic acid and other polyesters), and (3) Directly from Microorganisms (for example, Polyhydroxyalkanoates).

Bio-based fibers

Natural fibers can be characterized as bio-base filaments which can be utilized instead of traditional fiber-fortifying materials, for example, glass. The detail characterization of characteristic fiber is appeared underneath Fig. 1.

The *Animal* nature is loaded with precedents where human hair, chicken plume, sheep, alpaca, camels, rabbits and hairs of different winged creatures and creature are ordinarily depicted as a loss side-effect. Hair is a non-biodegradable waste accessible in ample quantity over the world, yet is not found completely for applications in designing field. Tensile strength of human hair

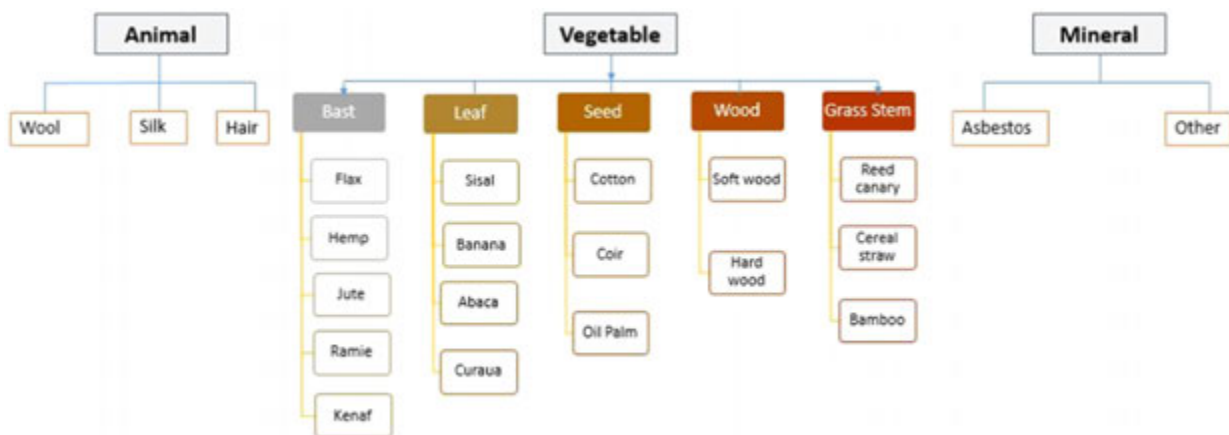


Fig. 1 Types of natural fiber.

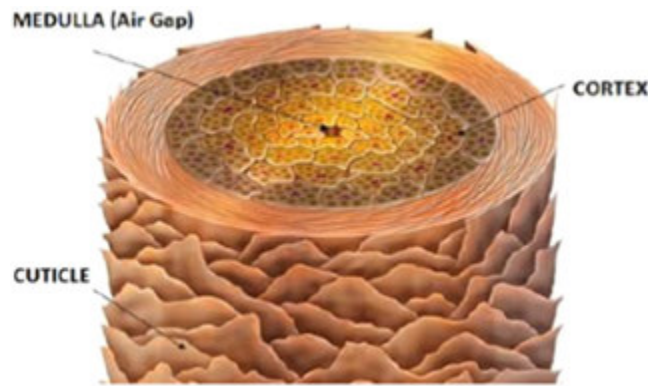


Fig. 2 Parts of hair fiber.

ranges from 150 to 220 MPa. While expanding the fiber volume portion up to a specific range the composite materials have demonstrated more noteworthy upgrades in their pliable properties (Fig. 2) (Rao *et al.*, 2017).

In this paper the emphasis is on the vegetable strands since all the others either have limited application or are absolutely denied by standard. Truth be told, creature filaments are not basically utilized and asbestos was prohibited because of dangers of presentation and dangers related with human wellbeing.

Vegetable fibers are found from the few pieces of the plants which are ordered into three fundamental gatherings relying upon the piece of the plant from which they are extracted.

Bast fibers

Bast fibers are defined as those obtained from the outer cell layers of the branches of various plants. The fibers find use in textile applications and are gradually being considered as reinforcements for polymer–matrix composites as they are observed to be “sustainable”. Bast fibers which is the costliest and luxurious within this category, are valued for their exceptional coolness and freshness in hot weather. These fibers tend to have high durability and their qualities improve with age and cleaning. It is also able to absorb a good depth of color in the dyeing process and remains more colorfast for a longer period.

Flax delivers strong and stiff fibers and it can be grown in moderate climates. It is stronger than cotton due to highly oriented molecular structure. Flax cell is highly companionable with the human cell thereby producing a compassionate effect on the human organism. Flax fabric is an outstanding filter protecting against a chemically aggressive medium, noise and dust. Flax used in bed, table, bath items for residential and commercial use as well as attire and technical products like luggage, bags, purses, sewing thread etc (Figs. 3 and 4).

Hemp is coarser and stiffer than flax and nearly around 3–15 feet long. Depending on processing, fiber may be creamy white, brown, gray, black, or green. It has high strength which makes it mainly suitable for cordage, twine & thread. It is used in shoes, hats, shirts, t-shirts, & jeans.

Jute is a solid however low extensible fiber principally because of composite like structure with profoundly situated long chain atoms. Jute fiber indicates great dampness ingestion limit because of essence of various polar-OH gatherings. Dampness recapture esteem might be up to 36% at 100% relative stickiness which is a lot higher than cotton. Jute fiber scorches and consumes without softening like cotton on warming to high temperature. Jute start temperature is around 193°C. The high explicit warmth esteem (1360 J/kg/K) results great warm protection of jute. It is used to produce carpet backing, rope, sugar and coffee bagging, cordage and twine (Figs. 5 and 6).

Ramie also known as rhea, grass cloth used for several thousand years in China. It is fast growing & can be harvested as frequently as every 60 days. Ramie fiber is ordered artificially as a cellulosic fiber, as are cotton, material, rayon and others. Fiber of ramie is entirely strong and has great elasticity. Its elasticity is eight times that of cotton and seven times more prominent than silk. Ramie fiber, positions first among every vegetable fiber in regard to quality and sturdiness. It has interesting normal for opposing decay when presented to climate conditions or submerged in water. Ramie fiber is outstandingly white and very glossy, surpassing blanched cotton and material individually. Like material and cotton, ramie has poor versatility and wrinkles effectively. It is used in many imported apparel item like sweaters, shirts, blouses, & suits.

Kenaf fiber has been generally utilized as support in composites in the course of recent years which is a most appealing option because of its fast development at various climatic conditions and guaranteeing ease; kenaf fiber has increased some thoughtfulness regarding supplanting the glass fiber composite and making it absolutely an eco-accommodating composite. Be that as it may, for improving its properties in various applications. A solitary fiber of kenaf can have an elasticity and modulus as high as 11.9 GPa and 60 GPa, separately. Same like jute, it is used for twine, cordage, & other technical purposes (Fig. 7).

To the extent composite applications are respected, flax and hemp are two strands that have supplanted glass in various parts, particularly in the German car enterprises.

- The elasticity of cloth string is twice as high as that of cotton and multiple times that of wool.
- Linen lessens gamma radiation about significantly and secures the human living being against sunlight based radiation.



Fig. 3 Fibers from Flax plant.



Fig. 4 Fiber from Hemp plant.

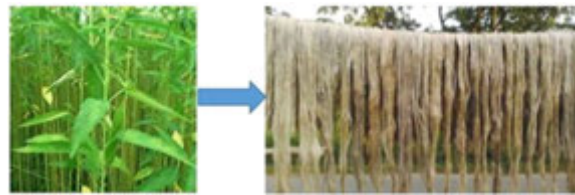


Fig. 5 Fiber from Jute plant.



Fig. 6 Fiber from Ramie plant.



Fig. 7 Fiber from Kenaf plant.

- According to therapeutic examinations led by Japanese analysts, wearing material garments disposes of some skin illnesses – from regular rash to perpetual skin inflammations.
- Linen is profoundly hygroscopic as it is proficient to quickly assimilate and yield dampness. That clarifies why material fabric dependably feels crisp and cool.
- Linen has high air penetrability and warmth conductivity properties. Warmth conductivity of cloth is five times as high as that of wool and nineteen times as that of silk.

Leaf fibers

Leaf filaments or hard strands are the hardest of the plant filaments which is doubtlessly because of their expanded lignin content when contrasted with different gatherings of plant strands. Application are ropes and coarse materials. Inside the complete generation of leaf fiber, sisal is the most important. It is acquired from agave plant. The firmness is moderately high and usually connected as fastener twines.

Sisal filaments are smooth, straight and yellow. It is utilized for better evaluations of rope, twine and brush bristles and despoiled by salt water. It is additionally utilized in mixes with wool and acrylic for a milder hand. Sisal is utilized for upholstery, divider covers and custom floor coverings gives intriguing surfaces to numerous inside styles (Mancino *et al.*, 2018; Lima *et al.*, 2014) (Figs. 8 and 9).

Abaca originates from individual from banana tree family. Its strands are coarse, extremely long up to 15 feet, solid, tough, and adaptable. It is grayish to dark colored in shading. It is utilized for ropes, cordage, floor mats, table materials, some wicker furniture, and clothing.

Bamboo fiber is a one of a kind biodegradable material. As a characteristic cellulose fiber it tends to be 100% biodegraded in soil by microorganisms and daylight. The decomposition process does not bring about any contamination in the earth (Fig. 10).

To the extent composite is concerned, sisal is regularly connected with flax in mixture mats to give great penetrability when the tangle must be impregnated with a resin. In some inside applications sisal is favored in light of its low dimension of smell contrasted with strands like flax.

Seed fibers

Cotton is the most widely recognized seed fiber and is utilized for material everywhere throughout the world. Other seed strands are connected in less requesting applications, for example, stuffing of upholstery (Figs. 11 and 12).

Coir is the fiber of the coconut husk, it is a thick and coarse however sturdy fiber. It is utilized in ropes, tangling and brushes (Verma *et al.*, 2013).

Sugarcane Bagasse (SCB) squanders are picked as a perfect crude material in manufacturing new items on account of its low manufacturing expenses and amazing green end material. The related expenses of extraction, synthetic changes and additionally

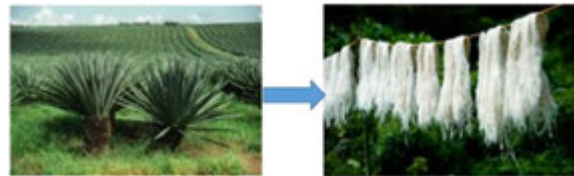


Fig. 8 Sisal fiber from Agave plant.



Fig. 9 Abaca fiber from Banana tree.



Fig. 10 Fiber from Bamboo tree.



Fig. 11 Fiber from Cotton plant.

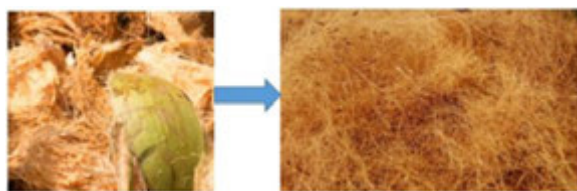


Fig. 12 Coir fiber from coconut husk.



Fig. 13 Bagasse fiber from Sugarcane.

Table 1 Properties of some vegetable fibers

<i>Plant Fiber</i>	<i>Cellulose (wt%)</i>	<i>Hemi cellulose (wt%)</i>	<i>Lignin (wt%)</i>	<i>Tensile strength (Mpa)</i>	<i>Young's Modulus (GPa)</i>
Flax	71	18–21	2	345–1035	28
Hemp	68	15	10	690	–
Jute	61–71	14–20	12–13	393–773	27
Sisal	65	12	10	511–635	9–22
Abaca	56–63	20–25	7–9	400	12
Bamboo	26–43	30	21–31	140–230	11–17

other pretreatment of SCB in the change procedure to prepared to-be utilized materials are possibly diminished as the mind bogging forms are disentangled by the simple use of Bagasse (Balaji *et al.*, 2014–2015; Loh *et al.*, 2013) (Fig. 13).

The fundamental arrangement and pertinent mechanical properties of the most widely recognized cellulose based filaments are appeared underneath Table 1 (Baptista *et al.*, 2013; John *et al.*, 2008; Balaji *et al.*, 2014–2015).

Fiber-Matrix Interface

The mechanical properties of fiber reinforced polymer composites are of incredible importance in choosing their end applications. Mechanical properties of composites rely upon the properties of the constituent strands, the network, and the fiber/framework interfacial shear quality. Inside the ascent of composite materials there is a reestablished enthusiasm for normal filaments. Their moderate mechanical properties limit the strands from utilizing them in innovative applications, however for some reasons they can contend with glass filaments.

Fabrication Process

Bio polymer composites created from normal strands is as of now the most promising region in polymer sciences. As a rule, just a single part of the composites, either the filaments or the resin, has been biodegradable. Such semi-green composites have a similar issue of transfer toward an incredible finish (Roy *et al.*, 2014).

The uncrushed regular strands were cleaned and substance treated utilizing Isocyanate, washing with basic arrangement, acrylic corrosive, and mercerization were connected. After this process, the common fiber was dried in an oven or air and for reduction in size it is processed in ball milled at 200–300 rpm for 5–6 h. The regular fiber and polymer framework were blending in the reactor or thermo kinetic mixer. After the blending composites were compressed under pressure from 6 to 10 MPa at 150°C to 170°C lastly dry the composites in dry air for appropriate relieving (Balaji *et al.*, 2014–2015) (Fig. 14).

There are three primary approaches to produce bio-base polymers;

- (1) By utilizing normally happening polymer through change, for instance cellulose subsidiaries, thermoplastic starch.
- (2) By creating bio-base monomers by maturation and polymerization, for instance Polylactic acid (PLA), bio-based nylon 6.

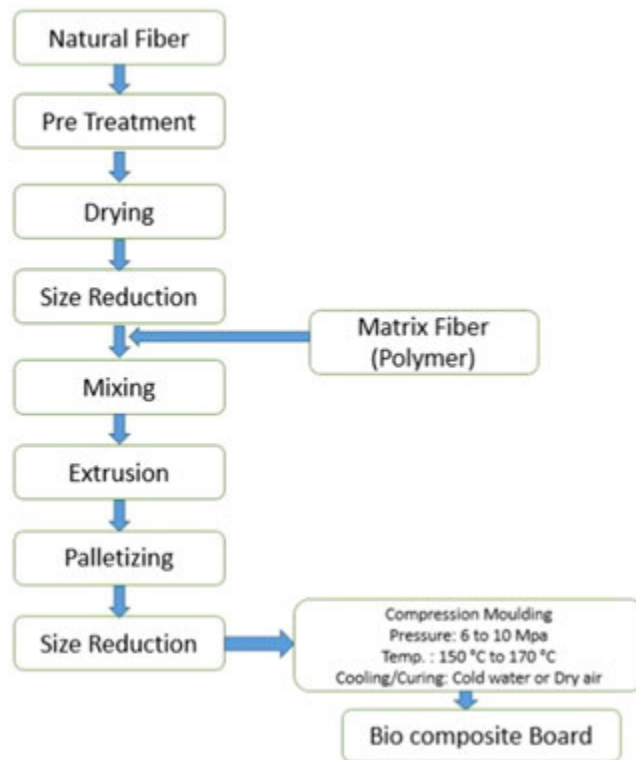


Fig. 14 Green Common Conversion of Bio composite processing.

- (3) By creating bio-based polymers with the assistance of hereditarily adjusted harvest as well as microorganism, for instance, poly hydroxyl alkanates.

There is an enormous open door in growing new bio based items, yet the genuine test is to plan reasonable bio-based items through advancement thoughts. To fabricate high quality composites, every one of the three factors to be specific fiber properties, resin properties as well as fiber/resin interface attributes are basic. The manufacture of super quality bio-composite has been portrayed in the plan (Fig. 15).

The manufacture of Kenaf fiber reinforced polypropylene sheets that could be thermoformed for a wide assortment of uses utilizing a pressure forming process using the layered filtering of a miniaturized scale fine polypropylene powder and slashed kenaf strands has additionally been finished. This creation technique has been used to make bio-composites utilizing a carding procedure, which is utilized to make uniform mixes of intermittent characteristic strands, for example, kenaf or jute with engineered filaments for use as the lattice (Roy *et al.*, 2014 (Fig. 16); Haydaruzzaman *et al.*, 2009; Yamamoto *et al.*, 2007).

Applications of Bio Composites

They are generally utilized for various applications as Automotive Industry, Aerospace Industry, Building Industry, Furniture Industry, Bio therapeutic Industry, and so on (Singh *et al.*, 2015; Gennusa *et al.*, 2017; Gunnal *et al.*, 2018; Ravikumar and Chandramohan, 2017).

Case Study in Modern Industry

Automotive industry

Natural fiber composites are being utilized for assembling numerous parts in the car division. The significant vehicle producers like BMW, Ford, and Toyota currently utilize characteristic fiber composites in a few applications as recorded in Table 2 (Hăloiu and Iosif, 2013).

AFT Plasturgie produces mixes of PP strengthened with hemp fiber in a level of 20%–40% for the car parts fabricating as appeared in Fig. 17 (Drzal *et al.*, 2001).

The European Industrial Hemp Association (Ashok Kumar *et al.*, 2017) deduced in their report, that all plastic strengthened with hemp fiber has points of interest like: low dimension of vitality required to create them, and furthermore a low dimension of

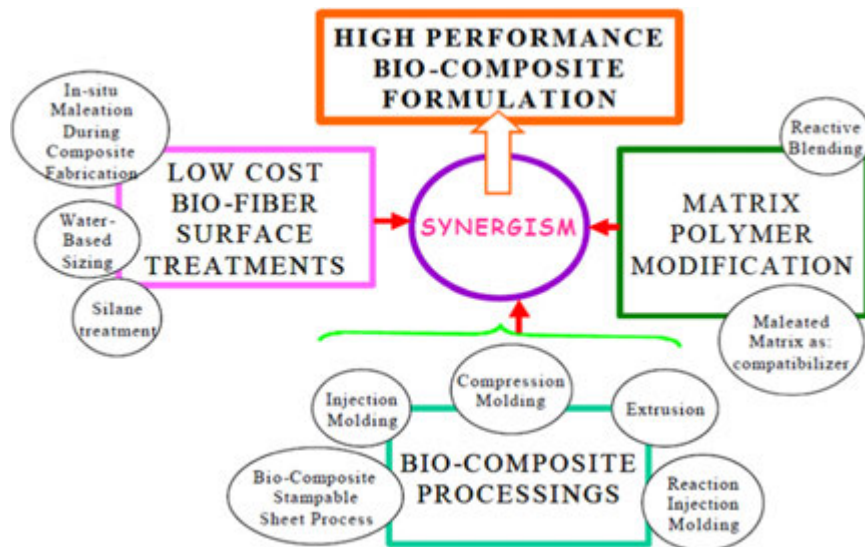


Fig. 15 Design of Superior strength bio-composites through ‘Synergism’ of Bio-fiber surface treatment, matrix polymer modification and Composite Processing. Adapted from Drzal, L.T., Mohanty, A.K., Misra, M., 2001. Bio-composite materials as alternatives to petroleum-based composites for automotive applications. *Magnesium*. 40 (60), 1–3.

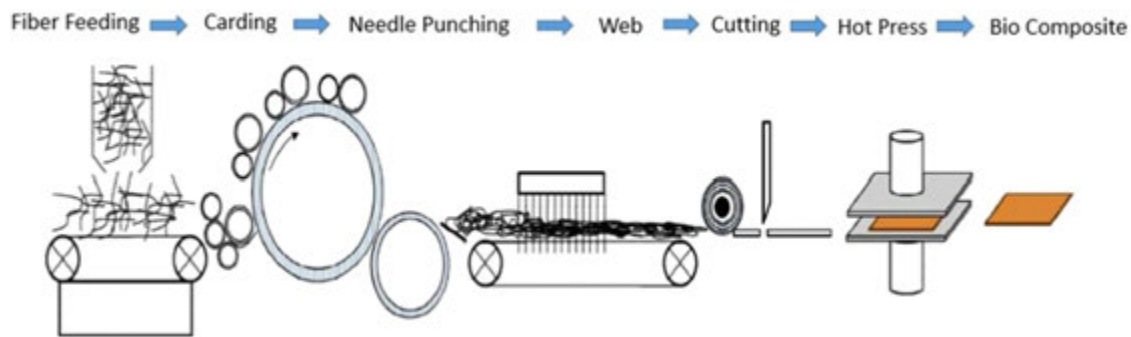


Fig. 16 Carding process for mixing natural fibers with Polypropylene Staple Fiber (PPSF).

Table 2 Percentage Reinforcement for various application of the bio source composite

Model	Fibers	Material	Application	Reinforcement (Bio Source)
BMW 7 series	Hemp	Acrylic polymer	Interior door panel	70%
Chrysler Sebring	Hemp, Jute	Polypropylene	Interior door panel	50%
Ford Fiesta and Focus	Jute	Polypropylene	Interior door panel	50%
The Ford Fusion and Lincoln	Soy	Polyurethane	Headrests	13%–26%
Nissan Leaf	Corn	Sorona	Rugs/Mats	20%–35%
Toyota	N.A.	Nylon bio-source	Cooling vessel	40%–60%

ozone harming substance outflows toward the finish of life, in examination with plastic reinforced with glass filaments. A case of a section acquired from bio-source composite is appeared in **Fig. 18** (Häloiu and Iosif, 2013; Kim *et al.*, 2011).

The idea of the work is to assess the practicality of some common fiber-strengthened arrangements and contrast with man-made materials. The investigation is broken into the following steps, the geometrical examination, the life cycle cost (LCC), and the life cycle assessment (LCA) (Khoshnava *et al.*, 2018; Herrmann *et al.*, 2014) (**Fig. 19**).

This segment outlines the consequences of various zone investigations. Life cycle engineering (LCE) is the craft of structuring the item under thought of the ecological and monetary effects related to specialized investigation. The Rocker and Hood parts are considering in this examination as appeared underneath figure (Hugo and de Carvalho, 2015) (**Figs. 20 and 21**).

Table 3 (Hugo and de Carvalho, 2015) shows Reinforcement properties of for Rocker and Hood Structure.



Fig. 17 Bio-source fiber reinforced parts produced by AFT Plasturgie.



Fig. 18 Door panel made of hemp fibers and epoxy resin.



Fig. 19 Scope of study.



Fig. 20 Rocker component in cycle.



Fig. 21 Bonnet component in car.

Table 3 Reinforcement properties for Rocker and Hood Structure

Constituent	E Glass	Carbon T300 (3K)	Rocker Part		Hood Part	
			Ramie	Jute	Ramie	Jute
Fiber Volume [%]	21	19.9	22.6	23.4	35.7	39
Fiber Mass [%]	36.5	26.8	27.1	26.3	41.4	42.7
Grammage [g/m ²]	300	193	190	180	300	300
E [GPa]	69	230	65	31.31	65	31.31
Poisson's ratio	0.25	0.2	0.3	0.38	0.3	0.38
Density [kg/m ³]	2600	1760	1530	1400	1530	1400
Filament diameter [m]	20	7	65.3	17.5	65.3	17.5
Lamina thickness [mm]	0.55	0.55	0.55	0.55	0.55	0.55
Tensile strength [MPa]	2750	3530	800	500	800	500

Table 4 Constituent composite comparative for Rocker Arm

Constituent	Thickness [mm]	N layers	Maximum Deformation [mm]	FOS
E-glass	19.80	36	1.70	1.96
CF T300	12.65	23	1.70	2.95
Ramie	19.80	36	1.70	2.24
Jute	24.75	45	1.70	2.11

Table 5 Constituent composite comparative for Bonnet component

Constituent	Max Deformation (mm)	FOS
E-glass	22.62	1.15
CF T300	21.59	1.50
Ramie	15.99	1.29
Jute	15.84	1.01

The final configuration for each fiber type is shown in below tables for Rocker arm and Bonnet components (**Tables 4 and 5**) (Hugo and de Carvalho, 2015).

Table 6 (Hugo and de Carvalho, 2015) show the comparison of weight among candidates for the same deformation.

The technical performance (TP) can be analyzed agreeing its stiffness [N/mm] and weight. The rocker segment firmness is straightforwardly connected with the solace and the steadiness. The aftereffects of every investigation are outlined in the **Table 7** (Hugo and de Carvalho, 2015).

In the specialized assessment two characteristics are considered, for example, the mass and the thickness of the Bonnet models. The deformation is dismissed since the design prerequisite for correlation motivations behind the models is accomplished. **Table 8** (Hugo and de Carvalho, 2015) summarizes the aftereffects of every investigation.

For the hood contextual investigation, the best decision is given between the carbon fiber, ramie and aluminum combination. Yet again, the ramie fiber is the best generally speaking when contrasting with E-glass and jute arrangements. Be that as it may, in the LCE affectability examination of the rocker, the best material decisions are the carbon fiber and the aluminum composite. And after that ramie, E-glass and jute, while considering just the fiber arrangements (Gulbarga and Burli, 2013).

Table 6 Weight per Constituent

Constituent	Model [g]		Fiber [g]		Matrix [g]	
	Rocker	hood	Rocker	hood	Rocker	hood
E-glass	163	2200	59	800	104	1440
CF T300	92	1550	25	420	67	1130
Ramie	136	1940	37	800	99	1140
Jute	170	2120	45	910	125	1210

Table 7 Rocker component, LCE

Material	TP [g]	LCC [Rs]	LCA [pt]
E-glass	163	1257.87	0.14007
CF T300	92	1229.36	0.14624
Ramie	136	1197.69	0.1221
Jute	170	1519.41	0.15193
Aluminum alloy	78	299.21	0.69196

Table 8 Bonnet component, LCE

Material	TP [g]		LCC [Rs]	LCA [pt]
	Mass (kg)	Thickness (mm)		
E-glass	2.20	4.40	4357.38	6.67
CF T300	1.55	3.30	6331.03	5.37
Ramie	1.94	4.40	3668.10	5.81
Jute	2.12	4.95	4266.53	6.28
Aluminum alloy	3.13	3.40	3498.07	11.63

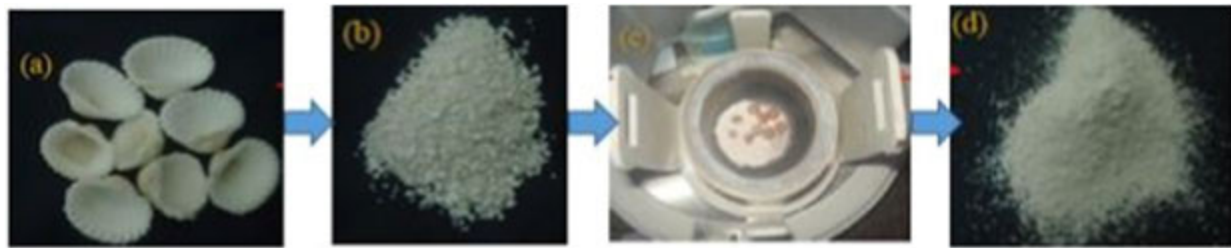


Fig. 22 Preparation of seashell nanopowder from sea shell.

Medical industry

The material utilized for manufacturing dentures ought to have great mechanical and tribological properties so as to withstand substantial powers inside the mouth. An investigation has been made to assess the hardness and tribological properties the Poly Methyl methacrylate (PMMA) based denture composite fortified with seashell Nano powder (Baptista *et al.*, 2013).

A denture is a removable swap for missing teeth and encompassing tissues. The significant property of such a denture is it ought to be perfect with the body environment. It must have great wear and mechanical properties so it could withstand overwhelming biting powers and wear created inside the mouth. The essential preparation of seashell nanopowder from ocean shell is appeared in Fig. 22. The seashell bio-material was at first grounded to littler particles by pounding and after that transferred to mono load ball-mill machine. The obtained nanopowder was then dried for 3 h so as to expel the humidity (Uyar *et al.*, 2016; Karthick *et al.*, 2014).

The readied examples were checked for smaller scale hardness under Vickers miniaturized scale hardness Tester (Karthick *et al.*, 2014) (Fig. 23).

The normal Vickers hardness (HV) of the examples are appeared in Fig. 24. It was discovered that the increase of seashell nanopowder demonstrated extended micro hardness.

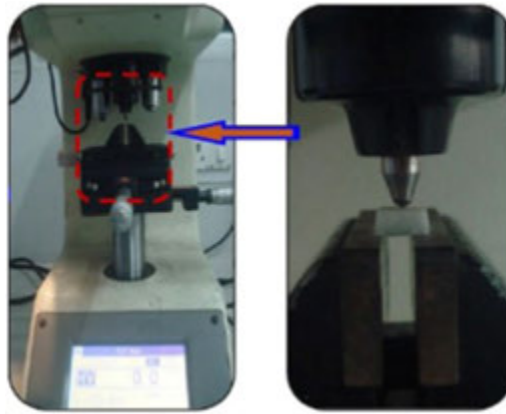


Fig. 23 Vickers micro hardness tester, BUEHLER, USA.

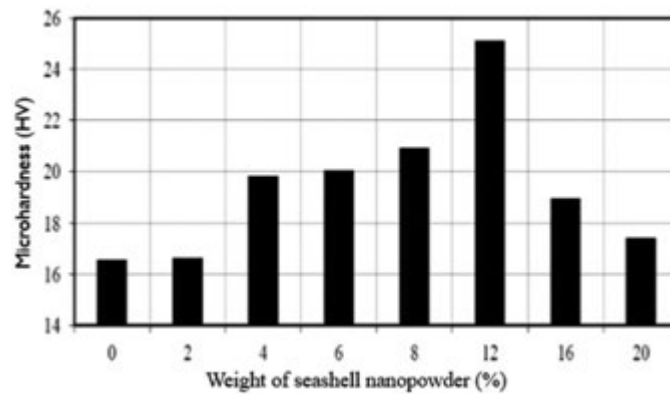


Fig. 24 Additional effect of seashell nanopowder on micro hardness values of the composites.



Fig. 25 Pin on disk apparatus, Ducom Instruments, Bangalore, India.

The readied examples were cleaned utilizing 600 coarseness sand paper in the Grinding machine. The example pursued ASTM standard G99–05. In wear tests, the specimens were made to slide against the hardened steel plate partner of hardness 954 HV with diameter of 100 mm and thickness of 6 mm. Diameter across of the wear track was 50 mm. After the specimen was mounted, switch arm was mounted by stabilizers of 15N. Wear try was performed for 15 min at a rotational speed of 40 RPM (Karthick *et al.*, 2014) (Fig. 25).

The expansion of 2% seashell nanopowder marginally expanded the miniaturized scale hardness esteems. The noteworthy change in the miniaturized scale hardness was seen when the seashell nanopowder content expanded to 4% and greatest smaller

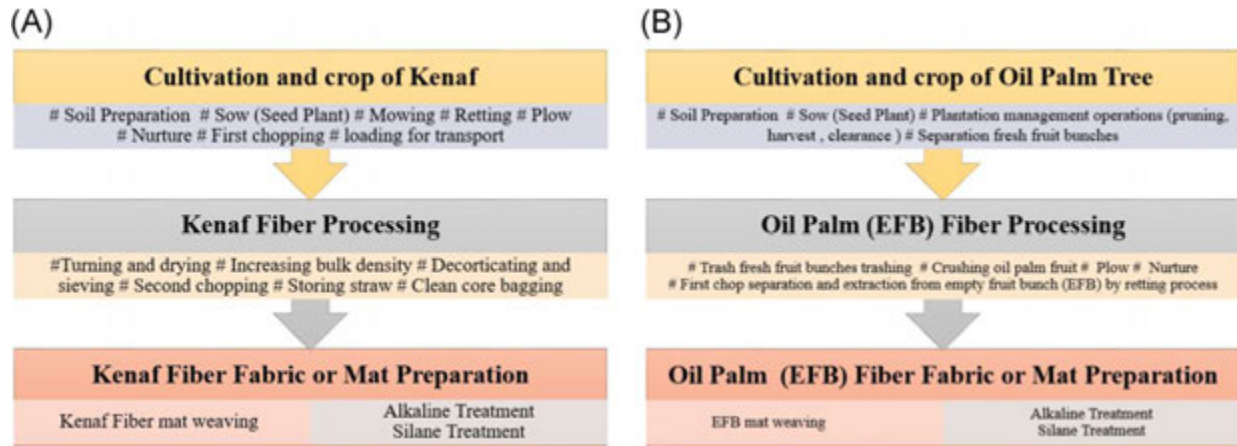


Fig. 26 Exemplary lifecycle of kenaf (part A) and oil palm EFB fiber (part B) mat preparation.

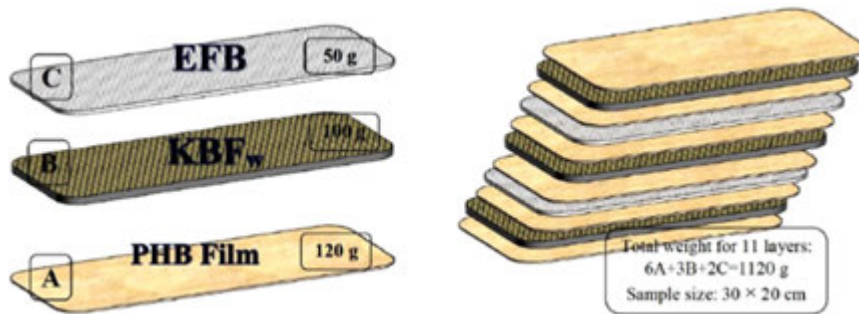


Fig. 27 Arrangement of KBFw/EFB hybrid PHB biocomposite.

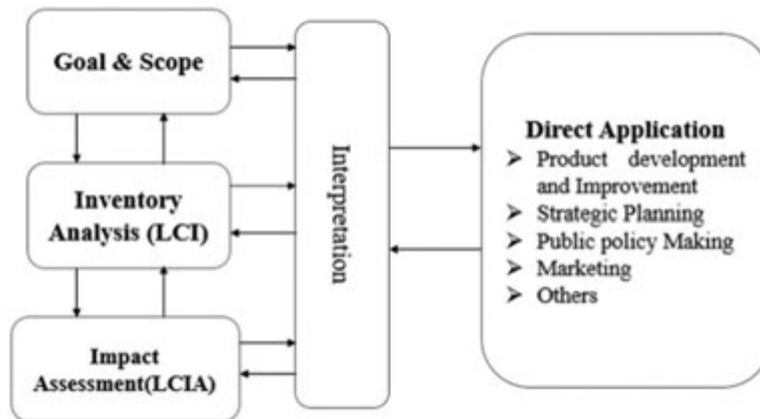


Fig. 28 LCA Phases (ISO 14040, 2006).

scale hardness was accomplished by including 12% seashell nanopowder. The uniform scattering and more grounded fortification in 12% filled composite was in charge of better wear property.

Construction industry

Adjusting the maintainability in development procedure can be accomplished through material choice procedure with low effect on condition and human wellbeing. Precisely, the kenaf as bast fiber is picked for this examination with predominant sturdiness and high perspective proportion in contrast with different strands as support in composite. Ecologically it has the most extreme carbon dioxide captivation among plants. The different properties among PHB (Polyhydroxybutyrate) and another two polymer PP (Polypropylene), and

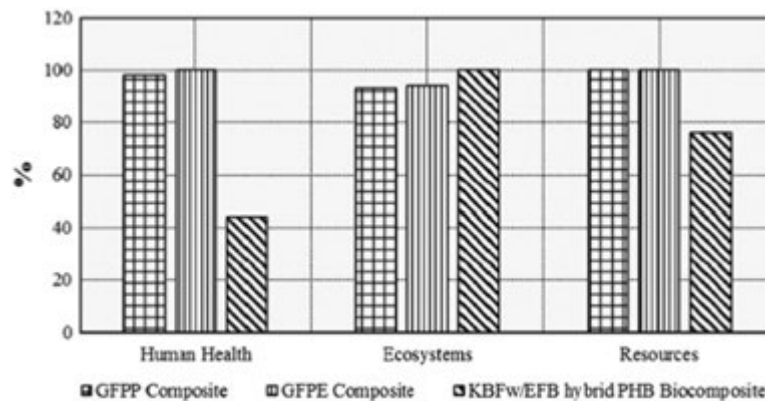


Fig. 29 Damage assessment factor of Impact assessment for 3 difference sample.

PE(Polyethylene) in light of Goodfellow site data. It thinks about noteworthy characters, and demonstrates some likeness in physical and mechanical properties of PHB, PP, and PE.

Composite material making from empty fruit bunches of palm oil (EFB). Among of every single Natural Fiber (NFs), the oil palm EFB filaments are hard and extreme that it has potential as support in composite applications (Khoshnava *et al.*, 2018 (Fig. 26) Grobman *et al.*, 2017; Stemmelen *et al.*, 2011; Zlatanić *et al.*, 2004a,b).

The Fig. 27 (Khoshnava *et al.*, 2018) demonstrates the example format of hybrid biocomposite with 11 layers' laminate (3 layers KBFw mat, 2 layers EFB mat, and 6 layers PHB film).

In this examination the primary point is to decide and look at the natural profile of crossover biocomposite with basic composite as unstructured structure materials. Under the ISO 14040 arrangement of measures, LCA comprises of four stages: objective and extension definition, life cycle inventory analysis (LCI), life cycle impact assessment (LCIA), and elucidation (Khoshnava *et al.*, 2018) (Fig. 28).

Portrayal markers for the endpoint elements of harm appraisal can be characterized as:

- (1) Human Health is depicted dependent on unit of years, the quantity of life lost years and the quantity of life crippled years.
- (2) Biological systems, considered the loss of species amid a specific time over a specific zone dependent on unit of Year.
- (3) Asset additional charges depends on up and coming asset generation over an infinitive time allotment (expecting steady yearly creation).

The harm appraisal markers are appeared underneath (Khoshnava *et al.*, 2018) (Fig. 29).

In this way, the aftereffect of this exploration gives significant judge mental data to policy creators and the forthcoming makers in the commercialization period of this new bio composites as structure materials (Zini and Scandola, 2011b).

Conclusion

This survey ought to be of an incentive to scientists who are keen on the best in course of biomaterial assessment and choice of biomaterials. The vast majority of Polymer Matrix Composite utilize manufactured fiber-materials as filler, however several sorts of engineered fiber materials can cause harm of the earth and the cost is costly. As a result of this unwanted properties, natural cordial and shoddy filler materials are looked to supplant the materials that have been utilized. Be-reason for these issues, biocomposites are increasing mechanical enthusiasm for a world concentrated on natural results (Vignesh *et al.*, 2014).

In Automobile Sector contemplates demonstrate that a vehicle weight decrease of 10% prompts an advantage of 3%–7% of fuel utilization and a decrease of 100 kg guarantees a declarations of CO₂ discharge with 10 g/km. For the hat contextual analysis, the best decision is given between the carbon fiber, ramie and aluminum composite. Yet again, the ramie fiber is the best in general when contrasting with E-glass and jute arrangements. It was decided in Medical Sector that Poly methyl methacrylate (PMMA) bio-composite could be effectively fortified via seashell nanopowder with better properties at 12% seashell nanopowder content pursued by 8% filled composite. Creating green or biocomposite as upcoming age of development materials, items, and procedures with the point of lessening the negative natural effects of development materials. Estimation of the ensuing effects from biocomposite (as green structure materials) life cycle for a boundless effect types, for example, worldwide environmental change, normal asset fatigue, ozone consumption, fermentation, eutrophication, human wellbeing, and Eco danger.

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Properties of Fiber-Matrix Interfaces of Natural Fiber Composites

Pedro J Herrera-Franco, Marista University of Mérida, Mérida, Yucatán, México and Scientific Research Center of Yucatan, Mérida, Yucatán, México

Alex Valadez-González, Scientific Research Center of Yucatan, Mérida, Yucatán, México

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Glossary

Fiber surface treatment Physical and/or chemical procedures used to modify the surface topography, morphology and chemistry of the natural fiber.

Fiber-matrix interphase The three-dimensional region formed by the contact between the fiber and matrix (the interface) but also incorporates the region of some finite thickness extending on both sides of the interface in both the fiber and matrix where physical, chemical and mechanical interactions take place.

Inverse gas chromatography Is a highly sensitive and versatile gas phase technique to study the surface and bulk properties of particulate and fibrous materials. A pulse or constant concentration of a known gas or vapor (probe molecule) is injected down the column at a fixed carrier gas flow rate. The retention time of the probe molecule is then measured by traditional GC detectors (i.e., flame ionization

detector or thermal conductivity detector). Measuring how the retention time changes as a function of probe molecule chemistry, probe molecule size, probe molecule concentration, column temperature, or carrier gas flow rate.

Micromechanical techniques Techniques used to probe the mechanical properties of the fiber-matrix interphase at the micro-scale level. They use single-fiber models and are based on common mechanical tests such as tensile loading and shear loading, compression loading.

X-ray photoelectron spectroscopy (XPS) Also known as Electron Spectroscopy for Chemical Analysis (ESCA), is used to determine the quantitative atomic composition and chemistry by irradiating a sample with monochromatic x-rays, and resulting in the emission of photoelectrons whose energies are characteristic of the elements within the sampling volume. An XPS spectra is created by plotting the number of electrons verses their binding energy.

Introduction

Because of the increasing awareness of environmental issues natural fibers have become important and there is a renewed interest in their use in the development of low cost “eco- friendly” natural fiber reinforced composites. Indeed, the mechanical properties of natural fibers containing cellulose in composite materials are the subject of current international research efforts (Fuqua *et al.*, 2012; Xie *et al.*, 2010; Thakur, 2015; Milanese *et al.*, 2011; Eichhorn *et al.*, 2001; Gholampour and Ozbakkaloglu, 2020). Natural fiber reinforced polymers have replaced many conventional metals/materials in various applications because they offer some advantages, such as the ease of processing, productivity, and cost reduction. In most of these applications, the properties of polymers are modified using fillers and fibers to suit the high strength/high modulus requirements. Fiber-reinforced polymers offer advantages over other conventional materials when specific properties are compared (Li *et al.*, 2007). Natural fibers have attracted the attention of scientists and technologists because of the advantages that these fibers provide over conventional reinforcement materials, and the development of natural fiber composites has been a subject of interest for the past few years. These natural fibers are low-cost fibers with low density and high specific properties; they are biodegradable and nonabrasive, unlike other reinforcing fibers such as engineering man-made fibers. Also, they are readily available and their specific properties are comparable to those of other fibers used for reinforcement (Herrera-Franco and Valadez-González, 2004).

In practice, there exist some drawbacks for the use of natural fibers as reinforcement of polymeric matrices, being the incompatibility between the naturally hydrophilic plant fibers and the hydrophobic polymer matrix the major one. Other factors also affecting the interfacial bonding between the fiber and matrix are the mechanical interlocking, the molecular attractive forces and the chemical bonds. In addition to the pectin and waxy substances in plant fiber acting as a barrier to interlock with the nonpolar polymer matrix, the presence of plenty hydroxyl groups hinders its operative reaction with the matrix. The tendency to form aggregates during processing, and poor resistance to moisture greatly reduce the potential of natural fibers to be used as reinforcement in polymers.

The fiber-matrix interphase can be considered as a three-dimensional reaction or diffusion zone in which two phases or components are physically, mechanically and/or chemically combined. Interfacial adhesion between the fiber and matrix plays a fundamental role in terms of the factors that govern the mechanical characteristics of the composite (Drzal, 1990). The overall mechanical properties of natural fiber reinforced polymer composites are highly dependent on the morphology, aspect ratio, hydrophilic tendency, fiber microstructure and dimensional stability. Several chemical treatments on cellulosic fibers that are used as reinforcements for thermosetting and thermoplastic polymers have been proposed (Belgacem and Gandini, 2005; Gholampour and Ozbakkaloglu, 2020). The modification of the surface characteristics of plant fiber and/or the hydrophobic polymer matrix is essential to formulate a reasonable composite with superior interfacial bonding and effective inherent stress transfer throughout the interphase. Various approaches, including physical treatments (i.e., solvent extraction, heat treatment, corona and plasma

treatments), physico-chemical treatments (i.e., laser, γ -ray and UV bombardment) and chemical modifications, including alkali, silane, acetylation, benzylation, acrylation and acrylonitrile grafting, maleated coupling agents, permanganate, peroxide, isocyanate, stearic acid, sodium chlorite, triazine, fatty acid derivate (oleoyl chloride) and fungal have been proposed for improving the compatibility and bonding between the lignocellulosic molecules and hydrocarbon-based polymers (Belgacem and Gandini, 2005; Li *et al.*, 2007). The significance of surface-treated natural fibers is seen through the improvement of mechanical strength and dimensional stability of resultant composites as compared with a pristine sample.

The increasing environmental awareness has brought substantial research and industrial invest in another class of plant fiber composites, in which the matrices including starch, cellulose, chitin and chitosan, collagen, lignin, natural rubber, poly-hydroxyalkanoate, polylactic acid (PLA) and soy-based resins, are fully biodegradable and sustainable biopolymers. Unlike synthetic polymer based composites, the similar polarities of both reinforcements and matrices impart the biodegradable composites better compatibility and interfacial adhesion (Mohanty *et al.*, 2005). However, some surface treatments of the fibers are specifically needed for the benefits of lowering down moisture sensitivity which generally leads to the dimensional instability due to the fiber swelling and hence loss of interface integrity.

The enormous potential in the field of recycled materials for civil construction in the use of waste natural fibers as reinforcement in cement matrix composites has been discussed by Savastano *et al.* (2005), Savastano and Agopyan (1998), and Tonoli *et al.* (2009). They found that, for mainly vegetable fiber composites, the transition zone is porous, cracked and rich in calcium hydroxide macro crystals. These characteristics have been directly related to the fiber-matrix bonding and to the composite mechanical performance. They also reported that the modification of the fiber surface showed significant influence on the microstructure of the composites (fiber-matrix interface and mineralization of the fiber lumen).

The characterization of fiber/matrix adhesion is of special importance for the interpretation of the mechanical properties of a composite. Different testing procedures have been proposed for the characterization of the natural fiber/matrix interactions, namely, the pull-out test, the microbond test and the single-fiber fragmentation test, (Graupner *et al.*, 2014; Rao *et al.*, 1991; Herrera-Franco and Drzal, 1992).

In this article, we revisit and summarize the earlier efforts to characterize the properties of the natural fiber/matrix interphase and its relevance in the control of final effective properties of a composite material. The physico-chemical fiber and matrix treatments will be described for polymeric and cementitious matrix material, next, the micromechanical experimental techniques to characterize the fiber/matrix interactions will be given and to conclude, a model system will be used to illustrate the fiber surface modification techniques, the physico-chemical characterization and a fiber-surface optimization study and its effect on the mechanical properties of a natural fiber polymer is presented.

Characteristics of Natural Fibers

Cellulose is the main component of natural fibers and the elementary unit of a cellulose macromolecule (Fig. 1) is composed of β -D-glucopyranose units which are linked together by (1 $\rightarrow$ 4)-glycosidic bonds (Gardner *et al.*, 2008). The length of a native cellulose molecule is at least 5000 nm corresponding to a chain with about 10,000 glucopyranose units. Cellulose molecules are linear and are aggregated through van der Waals forces and both intra- and intermolecular hydrogen bonds. Therefore all natural fibers are hydrophilic in nature (Sjöström, 1981).

Depending on their origin, natural fibers can be grouped into: bast (jute, flax, hemp, kenaf, mesta), leaf (pineapple, sisal, henequen, screw pine) and seed or fruit fibers (coir, cotton, oil palm). Unlike conventional man-made fibers like glass, aramid, carbon etc., that can be produced with a definite range of properties, natural fibers vary considerably. Cellulose of natural fibers contains different natural substances such as lignin and waxes. The fibers are made up of cellulose microfibrils bonded together by

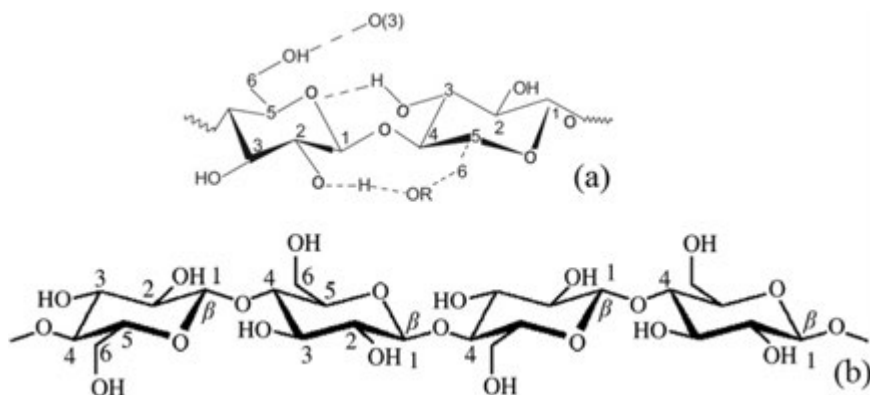


Fig. 1 Schematic drawing of: (a) a cellulose molecule; (b) intramolecular and intermolecular hydrogen bonds. Reprinted with permission from Herrera-Franco and Valadez-González (2005). Gardner, D.J., Oporto, G.S., Mills, R., Samir, M.A.S.A., 2008. Adhesion and surface issues in cellulose and nanocellulose. *Journal of Adhesion Science and Technology* 22, 545–567.

lignin, that is, they are a composite material by themselves. The physical properties of natural fibers are basically influenced by their physical and chemical structure such as cellulose content, degree of polymerization, orientation and crystallinity. In turn, these components are affected by the environmental conditions during growth of plants as well as the method used to extract them. Also, the fiber properties vary considerably depending on where they were taken from a plant, the plant quality and location. Different fibers have different lengths and cross-sectional areas and also different defects such as micro compressions, or pits or cracks (Rowland and Roberts, 1972; Cazaaurang-Martinez *et al.*, 1991). Another important parameter is the aspect ratio (length/diameter), which is highly modified by attrition during processing (extrusion, injection), has an influence on the mechanical properties of the composite (Dalvåg *et al.*, 1985).

It should be pointed out that cellulose swells when exposed to a polar media such as water, dimethylformamide, dimethylsulfoxide, tetrahydrofuran and pyridine. The hydroxyl groups are accessible for reaction, but some of these swelling solvents may be entrapped in the cellulosic structure (Bikales and Segal, 1971; Belgacem and Gandini, 2005). In contrast, non-swelling media (benzene, toluene, aliphatic hydrocarbons, etc.) force the hydroxyl groups to turn towards the inside of the cellulosic structure (Gauthier *et al.*, 1998). It is also possible, by progressive replacement of a swelling medium by media of lower and lower polarity, to maintain in nonpolar medium the initially swelled structure (Reichelt and Poller, 1981).

Polymer Matrix Composites

Composites of polymeric matrices reinforced with natural fibers is one major area of research. The study of polymer matrix composites has become an important topic for academic and industrial research. These composites exhibit moderate to good mechanical properties. Additionally, the current thrust for materials which are environmentally friendly and biodegradable made researchers to focus on alternate options to synthetic materials (Yashas Gowda *et al.*, 2018). Polymer matrix composites (PMC) are composed of various types of organic polymers reinforced with either short or continuous fibers with a variety of coupling agents which makes it possible to improve the mechanical properties such as fracture toughness, high strength and stiffness. The PMC are made in such a way that the applied mechanical loads are supported by the fibers. The function of the matrix is to keep the fibers together for an efficient transfer of load between them. Natural fiber reinforced polymer matrix composites could be cheaper, tougher and environmentally friendly, however the potential of such fibers for polymer composites has been the major subject of study during the last two decades (Cao *et al.*, 2005). Several approaches to improve the compatibility between the hydrophobic polymer and the hydrophilic fibers will be listed in this article.

Thermoplastic Matrix/Natural Fiber Composites

The use of thermoplastic polymers for engineering applications is common because of their excellent chemical resistance, good mechanical properties and lower cost. The major thermoplastic polymers used as matrix for composites are: Cellulose acetate, Nylon, Polystyrene (PS), Polypropylene (PP), Polyethylene (PE), Polycarbonate (PC), Polyvinyl chloride (PVC), Polyether-ether ketone (PEEK), Acrylonitrile-butadiene-styrene (ABS). Extrusion, injection molding and thermoforming are the most common manufacturing techniques. Fiber-fiber interactions as well as fiber-matrix interactions play a crucial role in determining the properties of such composites. It is observed that cellulosic fibers do not function as an effective reinforcement system due to poor adhesion at the fiber-matrix interface. Cellulose fibers also tend to aggregate and therefore the fibers do not disperse well in a hydrophobic polymer matrix and thus pose difficulties in achieving a uniform distribution of fiber in the matrix, (Nabi Saheb and Jog, 1999). Little attention has been paid to the fact that natural fibers are flexible and that depending on their initial length and the processing method used, their final shape will be distorted with shapes other than the stiff, straight fibers upon which all the micromechanical models were developed. Escalante-Solís *et al.* (2015) pointed out that because of the severe stresses exerted on the fibers during processing, it is very difficult to control directly their shape and orientation, but it can be done indirectly by improving the fiber-matrix interfacial adhesion, using fibers whose length is closer to the critical fiber length.

Thermosetting Matrix/Natural Fiber Composites

The biggest advantage of thermosetting polymers is their low viscosity making their introduction into fibers at low pressures very easy. Thermosetting matrix/natural fiber composites are processed by simple common processing techniques such as: hand lay-up, spraying, compression, transfer, resin transfer, injection, compression injection, and pressure bag molding operations. The major thermosetting polymers used as matrix for composites are: epoxy, phenolic, polyester, polyimide, polyurethane. Natural fiber-reinforced composites using a wide variety of natural fibers such as: kenaf, coir, coconut fiber, banana, sisal, jute, flax, vakka, and pineapple leaf have been fabricated with thermosetting polymer resins (Rao *et al.*, 2010; John and Thomas, 2008; Hepworth *et al.*, 2000; Joseph *et al.*, 1999). In thermosetting polymers, the fibers are used as unidirectional tapes or mats. These are impregnated with the thermosetting resins and then exposed to high temperature for curing to take place (Nabi Saheb and Jog, 1999). However, due to the inherent variations in the properties of natural fibers, process control at all possible intermediate steps is required to ensure reproducibility of end-use properties. Hence, proper drying of the reinforcement prior to fabrication is important (Ray and Rout, 2005).

Table 1 Natural fiber reinforced biodegradable polymer composites

Biopolymer matrix	Natural fiber reinforcement	References
Thermoplastic corn starch	Bleached <i>Eucalyptus urograndis</i> pulp Sugarcane and banana fibers Cellulose nanofibrils from wheat straw	Curvelo <i>et al.</i> (2001) Guimarães <i>et al.</i> (2010) Kaushik <i>et al.</i> (2010)
Thermoplastic rice starch	Cotton fiber	Prachayawarakorn <i>et al.</i> (2010)
Thermoplastic cassava starch	Cassava bagasse cellulose nanofibrils	Teixeira <i>et al.</i> (2009)
PLA	Jute and kapok fibers wood fiber Bamboo fiber and flour Kenaf fiber Flax Fiber Jute Fiber Banana Fiber Nanocellulose	Prachayawarakorn <i>et al.</i> (2013) Ding <i>et al.</i> (2016), Faludi <i>et al.</i> (2014) Wang <i>et al.</i> (2014) Ochi (2008) Bocz <i>et al.</i> (2014) Rajesh and Prasad (2014) Burrola-Núñez <i>et al.</i> (2019) Jandas <i>et al.</i> (2011) Song <i>et al.</i> (2014)

Note: Based on Zhou, Y., Fan, M., Chen, L., 2016. Interface and bonding mechanisms of plant fiber composites: An overview. *Composites Part B: Engineering* 101, 31–45. doi:10.1016/j.compositesb.2016.06.055.

Biodegradable Matrix/Natural Fiber Composites

A biodegradable biocomposite consist of a biodegradable polymer as matrix material and a bio-fiber as reinforcing material. The challenge in replacing conventional plastics by biodegradable materials is to design materials that exhibit structural and functional stability during storage and use and still remain susceptible to either microbial and/or environmental degradation upon disposal, without any adverse environmental impact. Biodegradable polymers may be classified as; biosynthetic, semi-biosynthetic and chemosynthetic. Such plastics can be made from renewable resources like starch, cellulose, etc. Examples of this type of plastics are starch plastics, cellulose acetate and polylactic acid (PLA) from corn. Natural fibers (lignocellulosics) are degraded by biological organisms since they can recognize the carbohydrate polymers in the cell wall. Lignocellulosics exposed outdoors undergo photochemical degradation caused by ultraviolet light (Nabi Saheb and Jog, 1999).

Zhou *et al.* (2016) has listed some current reports of the use of natural fibers to reinforce biodegradable polymers, and some representative works are reproduced in Table 1.

Cementitious Matrix/Natural Fiber Composites

The use of vegetable fibers such as sisal, jute and coconut in concrete poses an exciting challenge to the construction industry since they are a cheap and readily available form of reinforcement and require only a low degree of industrialization for their processing. When they are compared to the most common synthetic reinforcing fibers, the energy required for their production of equivalent weights, is small and therefore, their costs are also low (de Andrade Silva *et al.*, 2011). However, despite the enormous amount of experimental and analytical efforts dedicated to the mechanical characterization of the interface in man-made-fiber cement matrix systems, there is still a limited amount of data related to natural fibers (Peled and Bentur, 2003; Markovich *et al.*, 2001). Most of the interface characterization work has been performed on steel, glass and polymeric fibers. Naaman and Najm (1991), stated that there are four main factors that influence the bond between fiber and matrix: (1) physical and chemical adhesion, (2) mechanical component of bond such as deformed, crimped and hooked end fibers, (3) fiber-to-fiber interlock, and (4) friction. The tensile response of a continuous sisal fiber based cement and these distinct interface failure mechanisms are illustrated in Fig. 2.

The Fiber-Matrix Interphase

For the improvement of the interaction or interfacial adhesion of cellulose with hydrophobic materials it is possible to add a surfactant or to chemically modify its surface. The cellulose reactivity or the lack of reactivity depends on its structure. To modify cellulose structure, the highly ordered hydrogen-bonded lattice must be disrupted by swelling or dissolution. The reactive sites on cellulose, which may be derivatized, are the three-hydroxyl groups indicated as C-2, C-3 and C-6 (Fig. 1). C-6 is a primary hydroxyl, which is the most reactive position for esterification reactions while C-2 is the more acidic of the two secondary hydroxyl groups and is the more reactive site for etherification.

As a result of the modification of the cellulose structure and the adhesion with a hydrophobic material, a growing body of experimental evidence points to the creation or existence of a three dimensional region different in structure and composition near the fiber-matrix interface, i.e., an “interphase”. These results have led to an understanding of the inter-relationships between fiber, interface, and matrix, giving birth to the concept of the interphase, i.e., a three-dimensional region existing between the bulk fiber and bulk matrix (Drzal *et al.*, 1983). This interphase includes the two-dimensional region of contact between the fiber and matrix (the

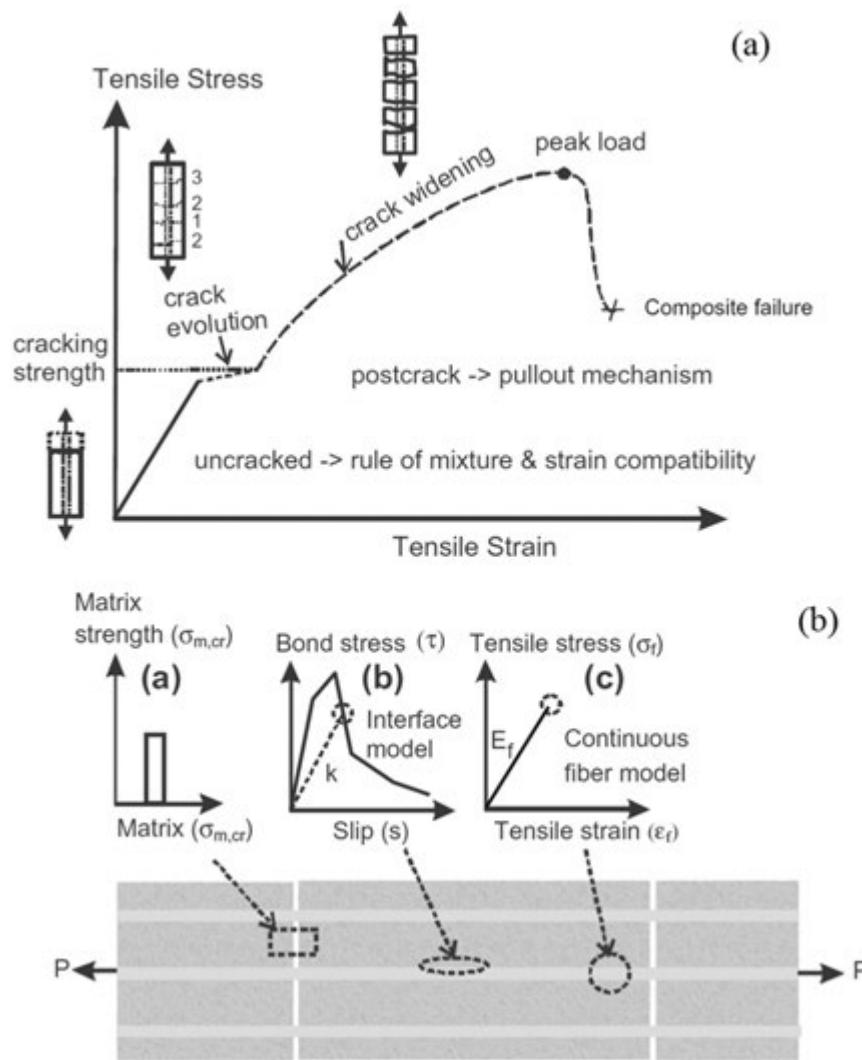


Fig. 2 (a) Schematic drawing of the tensile response of continuous sisal fiber-cement matrix composite; (b) Schematic drawing of a continuous sisal fiber reinforced composite, defined by three distinct mechanisms. Reprinted with permission from de Andrade Silva, F., Mobasher, B., Soranakom, C., Dias Toledo Filho, R., 2011. Effect of fiber shape and morphology on interfacial bond and cracking behaviors of sisal fiber cement based composites. *Cement & Concrete Composites* 33, 814–823. doi:10.1016/j.cemconcomp.2011.05.003.

interface) but also incorporates the region of some finite thickness extending on both sides of the interface in both the fiber and matrix. The “interphase” concept also allows for the inclusion of both interfacial as well as material mechanisms. For example, it has been shown that the fiber and matrix surface energy as well as the chemical bonding of the polymer on the fiber surface contribute to adhesion. Likewise, the material properties of the polymer near the fiber surface control the stress transfer and failure mode between the fiber and matrix. Furthermore, chemical and thermal shrinkage arises in specimens during cure and cool-down as well as from the differences in the mechanical properties of the constituents. These residual stresses that develop in the interphase can greatly affect the fiber-matrix adhesion (Gorbatkina, 1992). The complexity of the interphase can best be visualized from the schematic shown in Fig. 3. The desirability to develop structure-processing-property relationships incorporating the fiber matrix interphase increases the need to characterize the fiber-matrix adhesion and the interphase (Drzal, 1983) and (Broutman, 1969).

Several techniques have been proposed for the measurement of fiber-matrix adhesion level, either using a single-fiber model or a high-fiber volume fraction composite, both that have been subjected to the same processing or environmental exposures encountered either during manufacturing and fabrication or while in service. Then, processing effects, moisture and solvent sorption, thermal exposures and fatigue, could be properly evaluated for their effect on composite properties.

Fiber Surface Treatments

Several models to relate the properties of composite materials to the fiber-matrix interfacial behavior have been proposed. It has also been proven that a combination of both, mechanical principles with some assumptions about the level of fiber-matrix

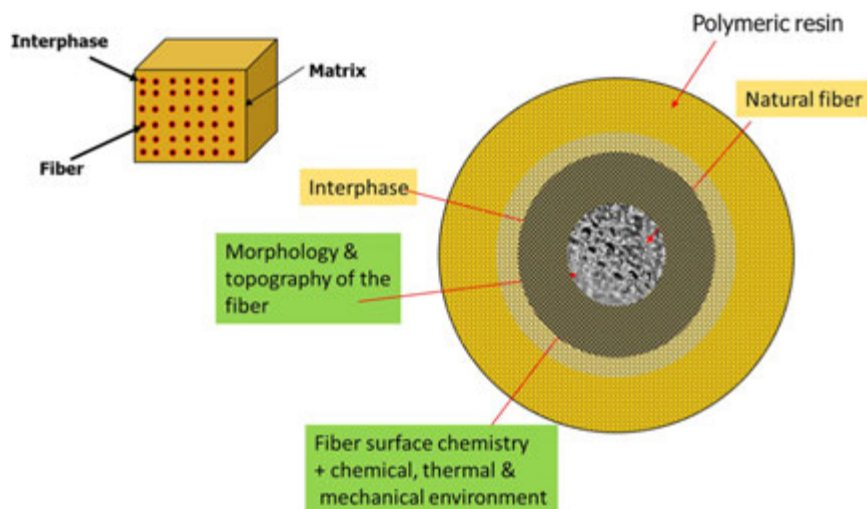


Fig. 3 Schematic of the concept of natural-fiber-matrix interphase.

adhesion and a modification of the fiber surface chemistry, e.g., tailoring of the fiber-matrix to control the final properties of the composite, appears to result in an optimum level of fiber matrix adhesion and the best mechanical properties (Valadez-Gonzalez *et al.*, 2009). In order to reduce the hydrophilic character of cellulose fibers and to improve the strength of their adhesion to the matrix, it is necessary to undertake a structural modification of their surface. Several approaches have been studied, namely (1) physical treatments such as corona, plasma, laser, Vacuum ultraviolet and γ -radiation treatments; (2) chemical grafting by direct condensation, including surface compatibilization with hydrophobic moieties and co-polymerization with the matrix. The copolymerization approach called upon different strategies: (1) The use of bi-functional molecules capable of reacting with the OH groups of the cellulose surface and leaving the second functions available for further exploitation; (2) The direct activation of the surface and the subsequent grafting from polymerization; and (3) The condensation of organometallic compounds, followed by their coupling with suitable reactive molecules or macromolecules (Belgacem and Gandini, 2005; Latif *et al.*, 2019; Koohestani *et al.*, 2019).

Physical methods

The physical methods of fiber surface modifications include the following: (1) physical treatments, such as solvent extraction; (2) physico-chemical treatments, like the use of corona and plasma discharges or laser, γ -ray and UV bombardment. Solvent extraction of lignocellulosic fibers is a simple procedure, mostly using a soxhlet, and as solvents: acetone, ethanol/toluene mixtures, diethyl ether and cold and hot water. The comparison of data from the X-ray photoelectron spectroscopy (XPS) spectra and Inverse gas chromatography (IGC) related to the pristine and treated fibers showed that at least part of the aliphatic and aromatic impurities (extractives and lignin fragments) originally present on the fiber surface were removed by these simple solvent extractions (Börås *et al.*, 1997; Belgacem *et al.*, 1995, 1996; Kamdem *et al.*, 1993).

Other physico-chemical treatments such as corona, dielectric barrier and plasma discharges and, more recently, laser, γ -ray and UV irradiations have been used to purify, oxidize and/or activate the surface of lignocellulosic fibers (Sawatari and Nakamura, 1992; Morales *et al.*, 2006). Discharge treatments such as low temperature plasma, sputtering and corona discharge are of great interest in relation to the improvement in functional properties of natural fibers. Low temperature plasma treatment causes mainly chemical implantation, etching, polymerization, free radical formation, crystallization, whereas sputter etching brings about chiefly physical changes such as surface roughness and this leads to increase in adhesion and decreases light reflection (Wakida and Tokino, 1996). The Corona treatment of one or both of the constituents of a compound also resulted in decreased melt viscosities relative to compounds containing untreated materials. These reductions were attributed to low molecular weight moieties formed on the surfaces of both polyethylene and cellulose during corona treatment which acted as lubricants at the fiber-matrix interface (Dong *et al.*, 1992).

Alkaline treatments

The alkaline treatment or mercerization is one of the most used chemical treatments of natural fibers when used to reinforce thermoplastic and thermosetting polymers (see Table 2). This treatment removes a certain amount of lignin, wax and oils covering the external surface of the fiber cell wall, depolymerizes cellulose and exposes the short length crystallites. Then, it can be said that the disruption of hydrogen bonding in the network structure results in an increase of fiber surface roughness, (Mohanty *et al.*, 2001). Addition of an aqueous sodium hydroxide (NaOH) solution to the natural fiber promotes the ionization of the hydroxyl groups to the alkoxide (Agrawal *et al.*, 2000):

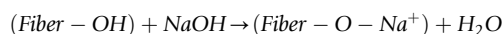


Table 2 Alkaline treatment of common plant fiber used in polymer matrix composites

<i>Fiber</i>	<i>Solution</i>	<i>Time & Temperature</i>	<i>Comments</i>	<i>Reference</i>
Henequen	2% aqueous NaOH solution	For one hour at 25°C	Increased mechanical interlocking	Valadez-Gonzalez et al. (1999a)
Jute	5% alkali (NaOH) solution	0, 2, 4, 6 and 8 h at 30°C	An improvement in the crystallinity, mechanical properties, water repellent behavior	Ray et al. (2001) , Varma et al. (1988)
Sisal	Alkalization: The defatted fibers were immersed in 5% NaOH solution.	1 h at 30°C		Mishra et al. (2001) , Orue et al. (2016)
Flax	NaOH at 3% and 10% 1500 mL of 10 g/L sodium hydroxide/ethanol solution	23°C, 60°C and 100°C for 1 and 2 h, 78°C for 2 h	Increased roughness, crystallinity and thermal stability	Cao et al. (2012) , Amiri et al. (2015)
Oil palm fibers	5% sodium hydroxide solution	48 h	Increased amount of amorphous cellulose at expense of crystalline cellulose	Sreekala et al. (1998)
Alfa fiber from <i>Stipha tenacissima</i>	NaOH at 1, 5% and 10%	0, 24, and 48 h at 28°C	Fibrillation	Rokbi et al. (2011)
Kenaf	5% 10% and 15% alkali solution.		Decrease of mechanical and physical properties.	Mahjoub et al. (2014) , Fiore et al. (2015) , Akhtar et al. (2016)
Borassus fruit fiber	5%, 10% and 15% NaOH		5% NaOH yielded improvement in tensile properties	Boopathi et al. (2012)

Note: Based on Kabir, M.M., Wang, H., Lau, K.T., Cardona, F., 2012. Chemical treatments on plant-based natural fiber reinforced polymer composites: An overview Composites Part B: Engineering 43, 2883–2892. doi:10.1016/j.compositesb.2012.04.053.

Then, an alkaline surface treatment directly influences the cellulosic fibril, the degree of polymerization and the extraction of lignin and hemicellulose compounds (Jähn *et al.*, 2002). In the alkaline treatment, the fibers are immersed in an aqueous NaOH solution for a given period of time. Henequen fibers were treated a 2% aqueous NaOH solution for h at 25°C, (Valadez-Gonzalez *et al.*, 1999a), jute and sisal fibers with 5% aqueous NaOH solution for 2 h up to 72 h at room temperature, (Ray *et al.*, 2001; Mishra *et al.*, 2001). Similar treatments were attempted by Morrison *et al.* (2000) to treat flax fibers.

Chemical treatments

The use of a fiber-matrix coupling agent is perhaps one of the most frequently used chemical surface treatments. Table 3, shows the most common silane coupling agents used to modify natural fiber surfaces for composites.

Experimental Methods for Interphase Characterization

A correlation between the mechanical behavior of the composite material under various loading conditions and the single-fiber composite failure mode during the fiber fragmentation test has been observed (Drzal, 1990). The critical parameters affecting the mechanical properties of a given fiber-matrix combination were identified to be the level of adhesion between the fiber and the matrix and the interphase morphology.

Micromechanics of Natural Fiber Reinforced Materials

For the interpretation of the mechanical properties of a composite, the characterization of fiber/matrix adhesion is of special importance. For the analysis of composites reinforced with man-made fibers, such as glass or carbon fibers, some micromechanical tests are already available (Narkis and Chen, 1988; Zhandarov and Mäder, 2005; Ji *et al.*, 2003; Müssig and Graupner, 2017; Graupner *et al.*, 2014; Zafeiropoulos, 2007; Herrera-Franco and Valadez-González, 2005). These tests are: the pull-out test, the microbond test (Gaur and Miller, 1989; Beckert and Lauke, 1997), the push-in test, the three-fiber test (Järvelä *et al.*, 1983) or the fragmentation test (van den Oever and Bos, 1998). Brief descriptions of these tests with their advantages and disadvantages can be found in Herrera-Franco and Drzal (1992). Also, a thorough discussion of the suitability of these methods for the measurement of the fiber/matrix interaction and the determination of the fiber/matrix adhesion of cellulose fibers in a PLA, MAPP or PP matrix was given by Graupner *et al.* (2014).

The pull-out and the microbond tests

In the pull-out test, the average interfacial shear strength (IFSS), is measured by pulling out the fiber of the matrix. To perform this test, a free end of the fiber is fixed to the load cell of a Minimat testing machine and the other end of the fiber is embedded in a block of a polymer matrix, (see Fig. 4). The force applied to the free end to pull it out of the matrix, is continuously monitored and recorded. Next, the apparent IFSS is calculated from the maximum force P , at which fiber debonding occurs using the following equation proposed by Kelly and Tyson (1965):

$$\tau = \frac{P}{\pi dL} \quad (1)$$

Here, d and L are the diameter and embedded length of the fiber. The critical fiber length L_c was calculated according to the equation proposed by Kelly and Tyson (1965) given in Eq. (2).

$$L_c = \frac{\sigma_f}{2} \left(\frac{d}{\tau} \right) \quad (2)$$

where σ_f is the tensile strength of the fiber.

The Microbond (or Microdrop) test (see Fig. 4) is another test proposed for the measurement of the IFSS. It is a variation of the fiber pull-out test (Miller *et al.*, 1987). The samples in this test are made depositing a droplet of the matrix resin on the fiber and once cured using the same conditions used to cure the composite material, then, the cured droplet is supported appropriately to apply a load and to debond it of the fiber. Successful shear debondings are obtained in most trials, and the nature of the recorded force curves during the trial distinguishes between proper shear debonding and slipping of the droplet or fiber breakage. The IFSS is also calculated using Eq. (1).

The single-fiber fragmentation test (SFFT)

The single fiber fragmentation test is also used to calculate the IFSS. A single fiber is embedded in a dumb bell shaped tensile specimen which is subjected to a tensile load, (Fig. 5). Upon loading, tensile forces are transferred from the matrix to the fiber, through the interphase. Depending on the level of fiber-matrix adhesion, that is, interphase integrity, tensile stresses build up in the fiber. At some stage, the fiber tensile strength is reached at points where there are imperfections and therefore, the stress concentrations are high enough and the fiber fractures at these points. This loading process is continued until the fiber-fragmentations reaches a saturation point and no more fiber-fracture occurs. The final fiber fragment length is referred to as the *fiber critical length*, l_c , which is a good indicator of the ability of the interphase to transmit loads between fiber and matrix. The ratio

Table 3 Some silane coupling agents used for natural fiber/polymer composites: chemical structures, organo-functionalities and target polymer matrices

Chemical Structure	Functionality	Abbreviation	Target Matrix	References
$\text{H}_2\text{N}(\text{CH}_2)_3\text{Si}(\text{OC}_2\text{H}_5)_3$ $\text{HN}(\text{CH}_2)_2\text{NH}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$	Amino	APS	Epoxy	Bisanda and Ansell (1991).
			Polyethylene Butyl rubber Polyacrylate PVC Polystyrene	Maldas <i>et al.</i> (1989) Abdelmouleh <i>et al.</i> (2005) Serier <i>et al.</i> (1991) Matuana <i>et al.</i> (1998) Maldas <i>et al.</i> (1989)
$\text{CH}_2 = \text{CHSiCl}_3$ $\text{CH}_2 = \text{CHSi}(\text{OC}_2\text{H}_5)_3$	Vinyl	VTS	Polypropylene Polyacrylate Elastomers, polyethylene, silicone elastomers, Unsaturated Polyester, Polyethylene, Polypropylene, Ethylene propylene diene monomer rubber (EPDM), Ethylene propylene rubber (EPR)	Maldas <i>et al.</i> (1989) George <i>et al.</i> (1996a) George <i>et al.</i> (1996b) Bengtsson and Oksman (2006) Raj <i>et al.</i> (1989) Nachtigall <i>et al.</i> (2007) Singh <i>et al.</i> (1996) Maldas <i>et al.</i> (1989) Abdelmouleh <i>et al.</i> (2007) Pothan <i>et al.</i> (2006) Abdelmouleh <i>et al.</i> (2007) Ismail (2003) Ismail <i>et al.</i> (2002) Beshay and Hoa (1990)
$(\text{RO})_3\text{Si} - (\text{CH}_2)_3 - \text{OOC}(\text{CH}_3)\text{C} = \text{CH}_2$	Methacryl	MPS	Polyethylene Polyester	
$\text{HS}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$ $\text{HS}(\text{CH}_2)_2\text{Si}(\text{OC}_2\text{H}_5)_3$	Mercapto	MRPS	Natural rubber PVC EPR, Polyurethane Rubber (PUR), Styrene-butadiene rubber (SBR), EPDM	
$(\text{RO})_3\text{Si} - (\text{CH}_2)_3 - \text{O} - \text{CH}_2\text{CHCH}_2\text{O}$	Glycidoxyl	GPS	Epoxy Butyl rubber Polysulfide	Bisanda and Ansell (1991) Abdelmouleh <i>et al.</i> (2005) Gassan and Bledzki (1997) Doan (2006)
$\text{R}_2 - \text{Si} - \text{Cl}_2$	Chlorine	DCS	Polyethylene PVC	Matuana <i>et al.</i> (1998) Pickering <i>et al.</i> (2003)
$(\text{RO})_3 - \text{Si} - \text{R}'' - \text{N}_3$ $\text{R}'' = -\text{C}_6\text{H}_4 - \text{SO}_2 -$	Azide	ATS	Polypropylene Polyethylene Polystyrene	Miller <i>et al.</i> (1988a) Miller <i>et al.</i> (1988b) McFarren <i>et al.</i> (1977)
$(\text{RO})_3\text{Si} - (\text{CH}_2)_{15}\text{CH}_3$	Alkyl	HDS	Polyethylene [53,65,66] Natural rubber	Abdelmouleh <i>et al.</i> (2007) Gliesche and Mäder (1995) Abdelmouleh <i>et al.</i> (2004)

Note: Based on Xie, Y., Hill, C.A.S., Xiao, Z., Militz, H., Mai, C., 2010. Silane coupling agents used for natural fiber/polymer composites: A review. Composites: Part A 41, 806–819. doi:10.1016/j.compositesa.2010.03.005. Nabi Saheb, D., Jog, J.P., 1999. Natural fiber polymer composites: A review. Advances in Polymer Technology 18, 351–363. Available at: [https://doi.org/10.1002/\(SICI\)1098-2329\(199924\)18:43.0.CO;2-X](https://doi.org/10.1002/(SICI)1098-2329(199924)18:43.0.CO;2-X).

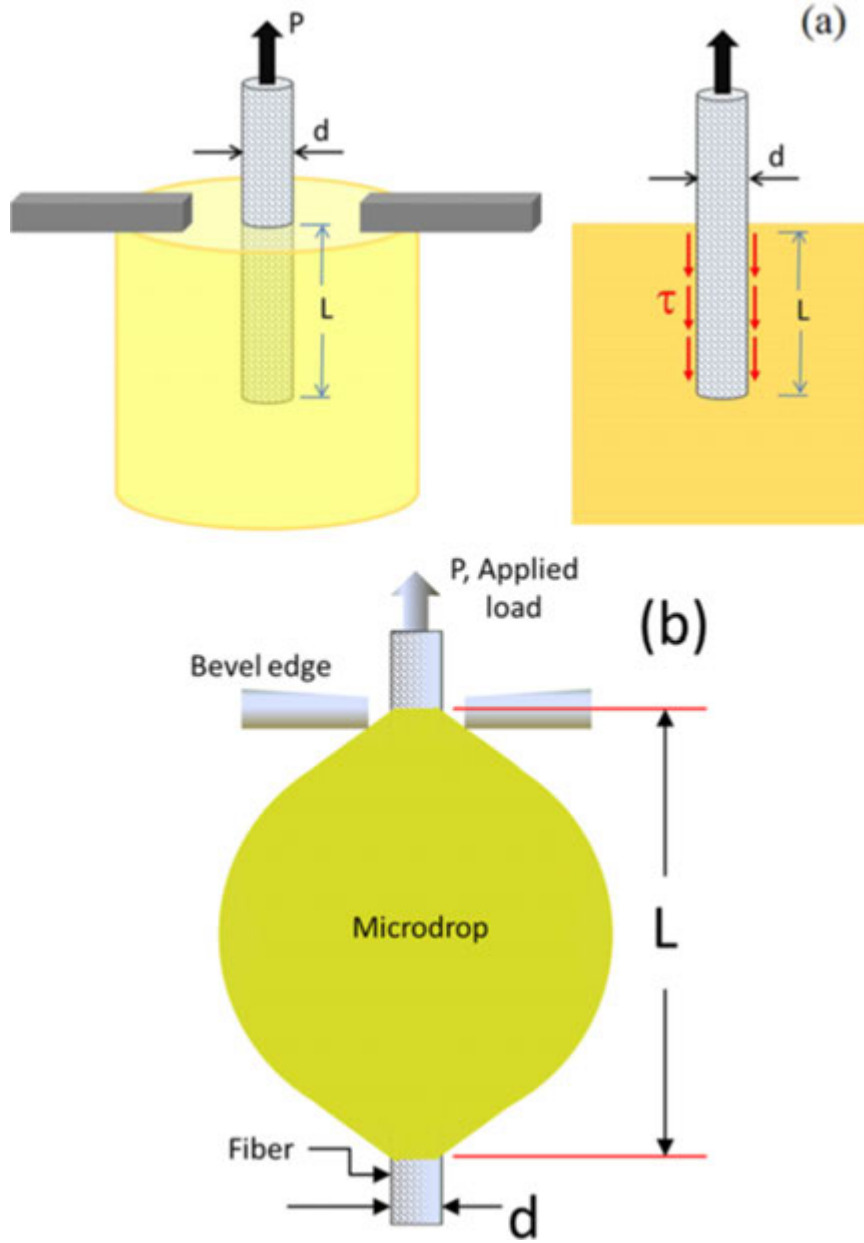


Fig. 4 Micromechanical techniques to probe the IFSS (a) Fiber pull-out; (b) Microbond (or microdrop).

(l_c/d) is also used as an indicator of the fiber-matrix bond strength. From the measured average value of l_c , the IFSS is calculated according to the equation developed by Kelly and Tyson (1965).

Drzal *et al.* (1980) recognized the random distribution of fiber fragments which is well described by a two-parameter Weibull distribution and rearranged the Kelly & Tyson equation proposing the following modification to calculate the IFSS:

$$\tau = \frac{\sigma_f}{2\beta} \Gamma\left(1 - \frac{1}{\alpha}\right) \quad (3)$$

Where Γ is the gamma function and α and β are the shape and scale parameters of the Weibull distribution respectively.

The inherent variability of the physical and mechanical properties of natural fibers like henequen and sisal has been amply documented in the technical literature (Cazaurang-Martinez *et al.*, 1991; Mukherjee and Satyanarayana, 1984; Barkakaty, 1976). Characterization of fiber mechanical performance is an exacting task with many potential pitfalls; however, it is intuitively obvious that most fiber testing will be least challenging experimentally when characterizing axial properties (Thomason, 2010). One of the characteristics that has been also documented is the fact that the cross-section and apparent diameter of these fibers vary considerably along their length, that is, they are not circular (Valadez-Gonzalez *et al.*, 1999a). Measurements of the average area and perimeter of the henequen fibers were performed from fiber segments embedded in an epoxy resin block. These fibers were subsequently sectioned at several points along the

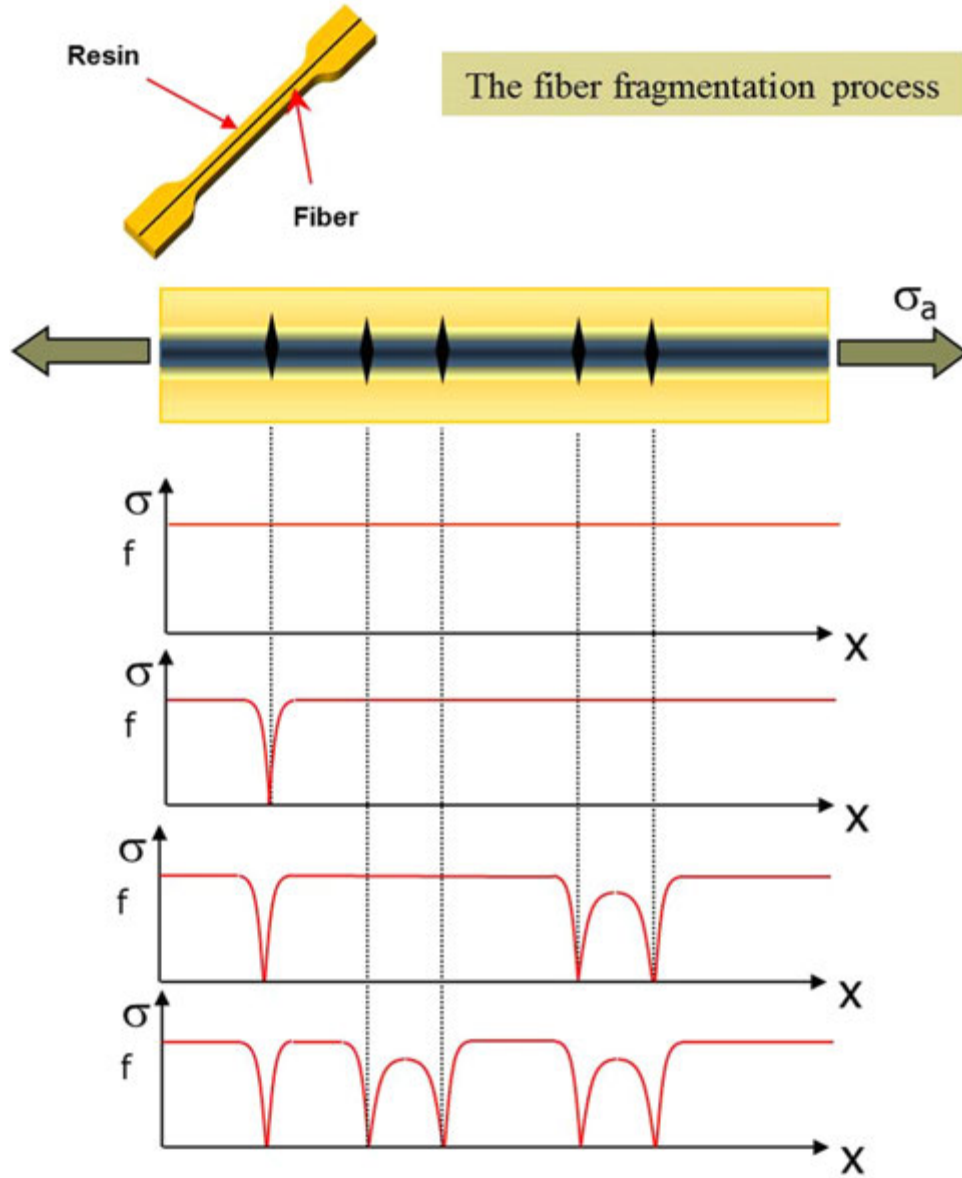


Fig. 5 Schematic representation of the single fiber fragmentation test showing the geometry of the specimen and the random nature of the fragment length distribution with increasing applied load.

fiber and the microtomed films were photographed to measure geometrical parameters of the fiber's cross-sections such as the perimeter and area, using commercial image analysis software. The variability of the perimeter, equivalent diameter and cross-section area of the henequen fibers, were fitted using a two-parameter Weibull distribution

$$F(x) = 1 - \exp\left\{-\left(\frac{x}{\beta}\right)^\alpha\right\} \quad x > 0 \quad (4)$$

The parameters of the Weibull distribution were estimated using the maximum likelihood method solving the following equations:

$$\frac{\sum X_i^\alpha \ln X_i}{\sum X_i^\alpha} - \frac{1}{\alpha} - \frac{\sum \ln X_i}{n} = 0$$

and

$$\beta = \left\{ \frac{1}{n} \sum X_i^\alpha \right\}^{\frac{1}{\alpha}} \quad (5)$$

Where X is the parameter being studied and α and β have the same meaning as indicated above. It was also observed that dispersion of the perimeter of the fiber cross-section is lower than that observed for the equivalent diameter or the cross-sectional area, thus,

suggesting that the perimeter could be a better parameter to calculate IFSS. Dong and Sapieha (1991) modified the Kelly & Tyson equation by using the ratio of the cross-section to perimeter of the fibers instead of the apparent diameter to calculate the IFSS when studying interfacial adhesion between cellulose and a thermoplastic matrix composite. In a study of the adhesion between polyethylene and regenerated cellulose, Karlsson *et al.* (1996) determined the fiber perimeters, using the Wilhelmy plate method. Model composites were prepared by embedding untreated and surface-fibrillated single fibers into an LDPE matrix, and the SFFT test was carried out to determine the critical fiber length. The interfacial shear strength (τ) was then calculated by applying a modified form of the Kelly-Tyson equation. In order to take into account the inherent variability of the henequen fibers, Eqs. (2) and (3) were rearranged to use the perimeter P_f instead of the diameter to calculate the IFSS. Following this approach the IFSS was calculated for the pull-out test with the expression (Valadez-Gonzalez *et al.*, 1999a):

$$\tau = \frac{P_d}{P_f l_e} \quad (6)$$

Where P_d is the force of debonding, P_f is the measured perimeter of the henequen fiber and l_e is the fiber embedded length. In the case of the single fiber fragmentation test the IFSS was calculated with

$$\tau = \frac{2P_b}{\beta P_f \Gamma(1 + 1/\alpha)} \quad (7)$$

Where P_b is the tensile failure load of the henequen fiber, P_f is the perimeter; α and β are the shape and scale parameters of the fragment length Weibull distribution and Γ is the gamma function.

Optimization of Mechanical Properties

Fiber-Physical and Mechanical Properties Versus Alkaline Treatment

As mentioned before, natural fibers are made up of cellulose microfibrils bonded together by lignin, that is, they are a composite material by themselves. Also, to overcome the issue of poor fiber-matrix adhesion, the natural fiber is subjected to an alkaline treatment removes a certain amount of lignin, wax and oils covering the external surface of the fiber cell wall, depolymerizes cellulose and exposes the short length crystallites. If additional treatments are applied to incorporate a coupling agent, acidic or basic mediums could be used in the process. As shown in Fig. 6, the henequen fibers were immersed in an aqueous NaOH solution (2% w/w) at ambient temperature, and the fiber-tensile strength shown as a function of gage length was affected by the severity of the surface treatment. There was a strength decrease of almost 20% after one hour of immersion. Therefore, a compromise has to be made between the time, temperature and alkalinity of the solution and the mechanical properties desired in the reinforcing material.

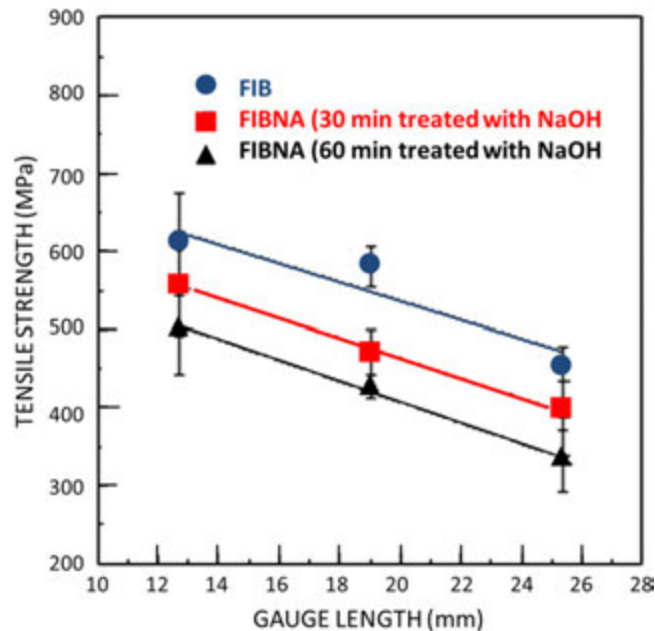


Fig. 6 Tensile strength of henequen fibers as a function of time of immersion in an alkaline solution.

Physico-Chemical Interactions at the Fiber-Matrix Interphase

In order to optimize the fiber surface treatment effects, a deep understanding of the interaction between the components in a composite system is necessary, then, the desired mechanism can be emphasized to promote bonding. This understanding will then allow the tailoring of the component surfaces to optimize composite properties for specific applications. As an example, the tensile and shear properties of henequen-high density polyethylene composite material were studied as a function of the concentration of the silane coupling-agent solution in order to optimize the composite properties. Also, the role of the silane coupling agent on the fiber-free surface energy is explored using Inverse Gas Chromatography (IGC) (Valadez-Gonzalez *et al.*, 2009).

Interfacial adhesion, wetting and mechanical properties of composites that depend on the interfacial characteristics are functions of the acid/base, or electron acceptor/donor properties of the materials involved (Schultz and Lavielle, 1989). The methodology of IGC is very convenient for the evaluation of acid/base interface potentials for a wide range of polymers and reinforcing fibers (Tshabalala, 1997; Coupas *et al.*, 1998; Papirer *et al.*, 2000). This allows the determination of surface characteristics of materials and it is a sensitive method to look at surface modification. Regarding cellulose fibers, IGC allows the determination of the dispersive component of surface energy using non-polar probes and acid base characteristics using polar probes.

When thermodynamic considerations are applied to IGC at infinite dilution, it can be shown that (Valadez-Gonzalez *et al.*, 2009):

$$-\Delta G_a = R.T. \ln V_n + k \quad (8)$$

where V_n is the retention volume of a vapor probe by stationary phase and k is a constant for a given chromatographic column. ΔG_a is related to the work of adhesion W_a by

$$\Delta G = N.a.W_a \quad (9)$$

Where N is Avogadro's number and a is the cross sectional area of the adsorbed vapor molecule. Combining Eqs. (8) and (9) leads to:

$$R.T. \ln V_n = NaW_a + \text{constant} \quad (10)$$

and since

$$W_a = 2(\gamma_s^d)^{1/2} a(\gamma_1^d)^{1/2} + C \quad (11)$$

$$R.T. \ln V_n = 2(\gamma_s^d)^{1/2} a(\gamma_1^d)^{1/2} + C \quad (12)$$

If $R.T. \ln V_n$ is plotted versus $a(\gamma_1^d)^{1/2}$, γ_s^d can be obtained from the slope of the linear function. That linear function can also be used as a reference for interaction with vapors that are able to interact with the substrate by non-dispersion forces. The distance between the retention volume for such a vapor and the reference line is the specific free energy change ΔG_{ab} due to polar interactions. Then,

$$\Delta G_{ab} = \Delta H_{ab} - K_d AN \quad (13)$$

If $\Delta G_{ab}/T$ is plotted versus $1/T$, ΔH_{ab} can be obtained from the slope of straight line. ΔG , ΔH and ΔS are the free energy, enthalpy, and entropy terms respectively. The enthalpy contribution may be expressed in terms of acid/base interaction as follows:

$$\Delta H_{ab} = K_a DN + K_d AN \quad (14)$$

Here AN and DN are the vapor phase acid/base characteristics and K_a and K_d are the acid/base interaction potentials of the solid. Then by dividing both sides by AN , Eq. (14) can be rewritten as

$$\frac{\Delta H_{ab}}{AN} = K_a \left(\frac{DN}{AN} \right) + K_d \quad (15)$$

The acid and base interaction parameters, K_a and K_d are readily obtained plotting $(\Delta H_{ab}/AN)$ versus (DN/AN) . The availability of K_a and K_d for components of a polymer system allows for an empirical evaluation of an acid/base pair interaction parameters, IP from Eq. (16)

$$IP = K_a(f)K_d(m) + K_a(m)K_d(f) \quad (16)$$

IP data are useful for the interpretation of the dependence of specific interactions of composite performance.

It has been reported from adsorption isotherms of the A-172 silane onto henequen fibers that a maximum value is noticed at a silane concentration of approximately 0.10% w/w and that at higher concentrations, the amount of silane adsorbed by the henequen fibers, decreases (Valadez-Gonzalez *et al.*, 1999b). They suggested that the formation of polysiloxanes inhibited this process.

A Particular Case of Fiber-Matrix Interphase Optimization

Material and Experimental Procedures

Materials

As a matrix, High density polyethylene (HDPE-Petrothene) extrusion grade, was supplied by Quantum Chemical Inc. with a melt flow index (MFI) of 0.33 gr/10 min, a density of 0.96 gr/cm³ and a melting point of 135 °C. Henequen fibers (Agave fourcroydes)

were supplied by DESFIYUSA. Sodium hydroxide and xylene, reactive grade from Técnica Química S.A., were used for the various surface treatments, and vinyltris (2-methoxy-ethoxy) silane (Silane A-172) from Union Carbide was used as coupling agent.

Henequen fiber surface treatments

The henequen fibers were treated with a NaOH aqueous solution (2% w/v) for an hour at 25 °C, and then washed with distilled water to eliminate the sodium hydroxide and then, dried at 60 °C for 24 h. The fibers were surface treated with 0.05%, 0.025%, 0.0125% and 0.00125% and 0.0067% w/w silane solutions using the following procedure: First, the silane was dissolved for its hydrolysis in a mixture of methanol-water (90/10 w/w), adjusting the pH of the solution to 3.5 with acetic acid with continuous stirring for 10 min. Next, the fibers were immersed in the solution and left for one hour under agitation and then dried first at 60 °C for 24 h. The fibers were maintained in the oven for 2 additional hours at 120 °C in order to assure the formation of covalent bonds between the fiber and the silane coupling agent (Valadez-Gonzalez *et al.*, 1999b).

Sample preparation

Pull-out test

To prepare the pull-out specimens, 20 cm long henequen fibers were placed between two sheets of HDPE and placed in a mold that was subjected to a 1 ton constant pressure, at 180 °C, using a Carver laboratory press for 10 min, and then cooled to room temperature. The specimens were cut in rectangles of 3 × 1 cm in such a way that a nominal fiber end was embedded. The embedded lengths of the fiber of each probe were recorded with a Specwell M820-SM optical microscope. The free end of the fibers were subjected to a tensile force while holding the polymer fixed using a Shimadzu universal testing machine Model A100 equipped with a 50 N load-cell and a cross-head speed of 1.2 mm/min was used in all the experiments.

The single fiber fragmentation test

The henequen single fiber fragmentation test specimens were prepared by carefully aligning and attaching the fibers to a frame and then, placed between two sheets of polyethylene and pressed at 180 °C in a Carver laboratory press for 10 min and then, cooled to room temperature. ASTM standard D-638 type IV specimens were cut using a Ceast Co. pneumatic hollow die punch, in such a way that one henequen fiber remained oriented along the central axis of the coupon. The specimens were subjected to a tensile load using an Instron 1125 Universal testing machine at 25 °C and a cross-head speed of 2 mm/min. The loading was stopped when the fragmentation process had ceased and before yielding of the polyethylene matrix. The fiber-fragment lengths were recorded with an optical microscope coupled to a calibrated TV camera and monitor system.

Elaboration of the composite

A composite with 20% fiber-volume fraction was elaborated to determine the effect of the henequen fiber surface treatments with the coupling agent (FIBNASIL) on the tensile and shear properties of the material and compared with composites made with untreated henequen fibers (FIB). Henequen fibers 6 mm long were incorporated to the polyethylene matrix using a BRABENDER intensive mixer at 180 °C; and laminated using a Carver laboratory press at the same temperature using a pressure of 1 ton. Type IV tensile test specimens were obtained according to the D638 ASTM standard. The tensile tests were carried out in an INSTRON Universal Testing machine model 1125 using a cross-head speed of 5 mm/min. The Iosipescu Shear Test was carried out following the ASTM D-5379 standard using a Wyoming Shear Test Fixture adapted to the Instron machine, after conditioning at 25 °C and a cross-speed of was 0.5 mm/min was used. The test specimens were cut to the following dimensions: 76 mm of length, 19 mm of width and 2 mm of thickness and the distance between the two 90° notches was 12 mm.

Inverse gas chromatography

The untreated henequen fibers (FIB) and the fibers treated with an aqueous NaOH solution followed by a 0.0125% silane solution (FIBNASIL) were analyzed in a Hewlett Packard HP 5890 Gas Chromatograph equipped with a flame ionization detector and an automatic injector. The retention time of the apolar and polar probes was measured at three temperatures (80 °C, 90 °C, and 100 °C). Dispersive energies were calculated using the increment of the free energy of a methylene group in the n-alkanes series (C₆ to C₁₀). The acid-base (specific) interactions were determined using the retention data of different polar probes: a basic probe, tetrahydrofuran (THF); an acid probe, Chloroform (CHCl₃); two amphoteric probes, Acetone (Ac) and diethyl ether (Ether). Methane was used as non-interacting reference probe. The properties of the probes used in this study are described in Valadez-Gonzalez *et al.* (2009).

Dispersive component of surface energy

It is known that the surface treatments modify the fiber surface energetic and so the nature of the fiber-matrix interphase. As shown in Fig. 7, the dispersive energy γ_s^D of the raw henequen fiber (FIB) and the silane treated one (FIBNASIL) decreases with the silane treatment for different temperatures. This decrease is an expression of an entropic contribution to the surface free energy. Other factors could include the increasing kinetic energy of the molecules with increasing temperature and possible changes on the solid surface. This makes the non-specific adsorption difficult, thus reducing the dispersive component of the surface energy. A similar behavior has been reported in other studies concerning lignocellulosic fibers. Price

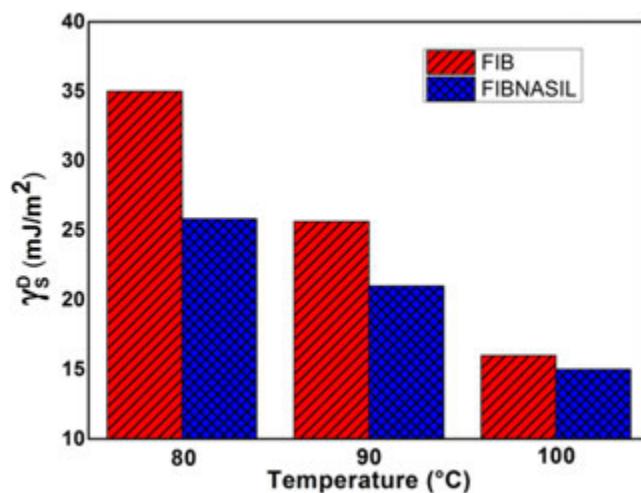


Fig. 7 Dispersive energy component γ_s^D for henequen-fiber surface for different temperature values. Based on Valadez-Gonzalez, A., Moreno-Chulim, M.V., Herrera-Franco, P.J., 2009. Modification of the fiber surface for the optimization of mechanical properties in natural-fiber reinforced polymers. *International Journal of Materials and Product Technology* 36, 417–430. doi:10.1504/IJMPT.2009.027846.

et al., using IGC found that the dispersive component of raw sisal fibers and alkaline treated ones were 24 ± 3.8 (Santos *et al.*, 2001; Price *et al.*, 2007).

One of the most popular semi-quantitative methods used for the evaluation of polymer surface acid-base characteristics involves calculation and comparison of their acceptor and donor constants (K_A and K_D) using Eqs. (13), (14) and (16). This method gives a gross indication of acid-base character, and has been used by several investigators for characterization of solid surfaces (Saint Flour and Papirer, 1982, 1983; Schultz and Lavielle, 1989; Felix and Gatenholm, 1993, Felix *et al.*, 1993; Kamdem *et al.*, 1993; Belgacem, 2000). At infinite dilution, with the probe concentration becoming zero in the solid phase, the interaction between the probe and the surface can be taken as a reversible physical exchange.

As shown in Fig. 8, with an additional specific component in the interaction, the values lie above the alkane reference line on the $RT \ln(Vn)$ versus $a(\gamma_1^d)^{1/2}$ plot. In Fig. 8(a), the tendency of the FIB surface to accept or donate an electron in an interaction is represented by the n-alkane straight line and is zero at the line with only dispersive interactions.

Furthermore, depending upon the nature of the polar probe molecule, the solid surface accepts or donates electrons and therefore, the polar probes lie above the n-alkane line (having dispersive and specific interactions). It can be noticed that chloroform, which is a Lewis acid probe, is lie near the alkane line, whereas THF and Ether, both Lewis base, lie far above the reference line. This is an indication of a greater interaction of THF and Ether with the FIB surface compared to CHCl_3 . On the other hand, acetone, an amphoteric probe, lies well above the reference line, indicating the greatest interaction with FIB. This behavior suggests that the FIB surface possess both acidic and basic sites. This result is not surprising at all if it is remembered that the henequen fibers are constituted of cellulose (70% w/w), hemicellulose (15% w/w), lignin (8%) and extractives (2%). The surface acid-base characteristics of cellulose and lignin are different. While cellulose is strongly acidic, lignin is more evenly bipolar, with a much weaker acidity and a similar basicity than cellulose. Then, the presence of a large number of ether-oxygen functional groups and a small number of acidic groups (e.g., phenolic and carboxylic hydrogen) in lignin are responsible for the weak acidity of lignin. The acetyl groups in the hemicelluloses also contribute to the basic character of FIB.

In Fig. 8(b) it is observed that the tendency of the FIBNASIL surface to accept or donate an electron in an interaction is different to that observed for FIB. It can be noticed that no matter what the specific character of all the probes are, they all lie closer to the reference line, indicating a lower interaction with them as compared to FIB. The Lewis acid probe, CHCl_3 lies at an equal distance as the basic probes, and ether and THF, whereas the amphoteric one is far away. This all suggests that the silanization of the henequen fiber stabilized the surface against specific interactions (Valadez-Gonzalez *et al.*, 2009). The specific interaction capability of the henequen fiber surface can also be represented through Lewis acid-base characteristics, reported in terms of the electron acceptor-donor capabilities. Using the temperature dependency of ΔG_{ab} given in Eq. (15) and using an approach mentioned by Schultz, a plot can be generated, as shown in Fig. 9, which readily gives the K_a and K_d in the form of the slope and the intercept, respectively.

It can be seen that the effect of the silanization reduces both the acid and base characters of the henequen fiber surface. However, it is also observed from the K_a/K_d ratio that the silane coupling agent increments the overall acidic nature of the surface (see Fig. 10). Such behavior can be attributed to the acidic silicon atom (Harding and Berg) or the uncondensed silanols (Si-OH) present in FIBNASIL surface.

X-ray photoelectron spectroscopy (XPS)

X-ray photoelectron spectroscopy (XPS) can provide information about surface composition and the chemical environment and bonding of surface chemical species. In the Fig. 11(a) and (b) the carbon peaks for FIB and FIBNA are presented. It can be seen that

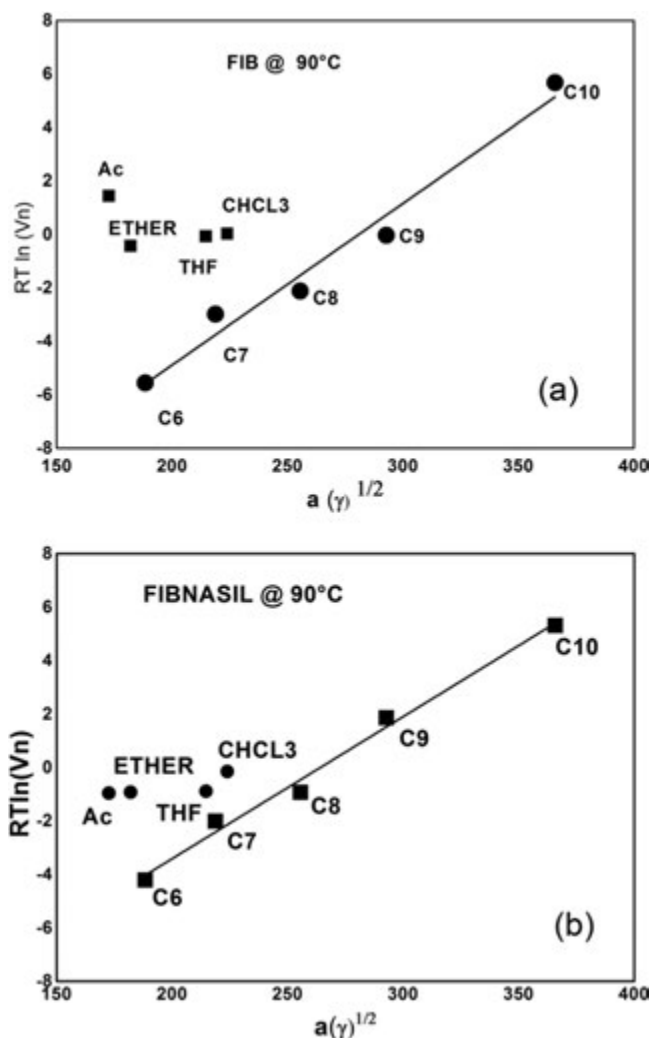


Fig. 8 Determination of ΔG_{ad} by the Schultz's method for: (a) FIB and (b) FIBNASIL at 80°C. Reprinted with permission from Valadez-Gonzalez, A., Moreno-Chulim, M.V., Herrera-Franco, P.J., 2009. Modification of the fiber surface for the optimization of mechanical properties in natural-fiber reinforced polymers. *International Journal of Materials and Product Technology* 36, 417–430. doi:10.1504/IJMPT.2009.027846.

for FIB there are three peaks at 288, 285.7 and 285.5 eV, respectively, indicating that there exists more than one compound on the surface of the fiber without treatment. Noah and Prud'home (1994), in a study of characterization of the surface of a tropical wood by XPS, also reported several emission peaks for the carbon atom and they attributed them to the different constituents such as, polysaccharides, lignin and waxes that are present on the surface of wood, each of them containing carbon atoms in different chemical environments. As mentioned, lignocellulosic fibers contain cellulose, lignin and waxes, the appearance of multiple peaks of emission for the carbon atom is expected and the treatment with the aqueous alkaline solution eliminated waxes and part of the lignin from the surface of the fibers. In Fig. 11(b) there is a slight shift of the peaks and the peak at 285.5 is no longer observed. The carbon peaks for FIBNASIL are shown in the Fig. 11(c). The effect of the silane surface treatment is reflected in the emission of a carbon peak for FIBNASIL, at shoulder at approximately 288 eV. This peak indicates the presence of the C–O–Si bond on the fiber surface and it means that a condensation reaction between the silane and the henequen fiber may have taken place. Fig. 11(d) shows the characteristic 2p and 2s emission peaks for silicon for FIBNASIL. In FIBNASIL it can be observed that there is a pair of peaks related to the 2p electrons, one of them approximately at 102 eV and the other close to 104 eV. The apparition of emission peaks at binding energies greater than 102 eV reflects the bonding of the silicon atom with more than two oxygen atoms. The coupling agent used in this study, has three ethoxy-methoxy groups attached to the silicon atom, so the hydrolyzed silane could have three hydroxyl groups attached to the Si atom. If this is true, then a peak should be expected at a binding energy greater than 102 eV. These findings seem to confirm the presence of the silane group deposited on the surface of FIBNASIL fibers and that there is a chemical reaction between the hydrolyzed silane and the henequen fibers.

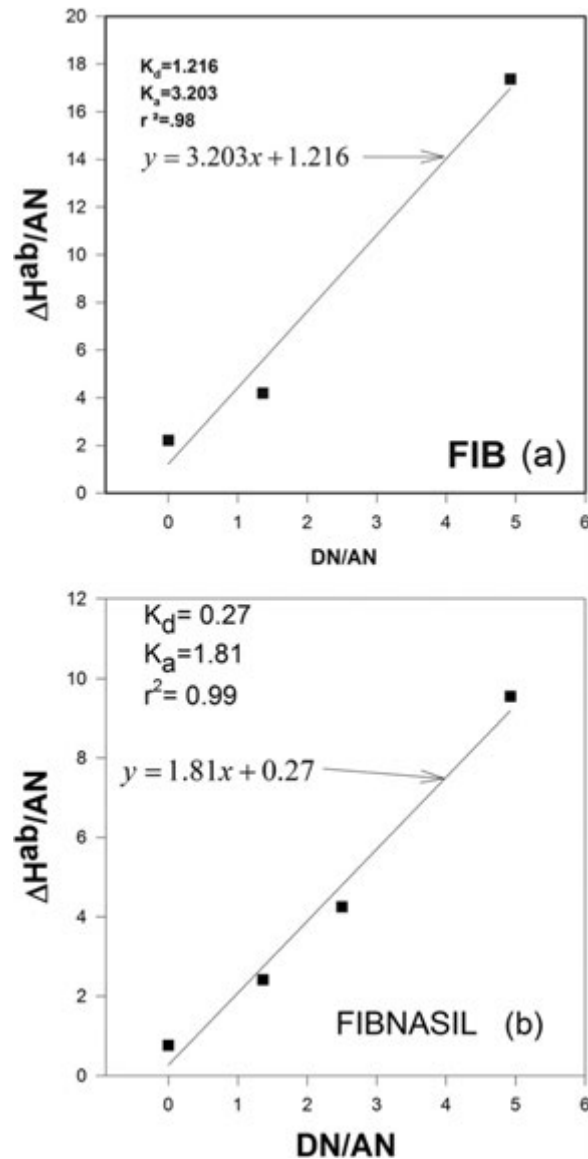


Fig. 9 Plots of $\Delta H^{ab}/AN$ as a function of DN/AN for the determination of K_a and K_d for (a) FIB; (b) FIBNASIL. Reprinted with permission from Valadez-Gonzalez, A., Moreno-Chulim, M.V., Herrera-Franco, P.J., 2009. Modification of the fiber surface for the optimization of mechanical properties in natural-fiber reinforced polymers. *International Journal of Materials and Product Technology* 36, 417–430. doi:10.1504/IJMPT.2009.027846.

In order to better appreciate the effect of the different fiber surface treatments on the composites behavior, tensile strength and Iosipescu shear strength testing results as a function of the silane concentration for a (80:20 v/v) composite (HDPE–henequen fibers) are shown in Fig. 12. A maximum of the amount of silane adsorbed on the fiber surface is observed, at a concentration of the silane coupling agent solution of approximately 0.0125%. The IFSS also has a maximum value at approximately 0.0025%. Similarly, the tensile strength attained a maximum values at a silane concentration below 0.0125%.

It is evident from the values of the mechanical properties of the composite that, the total amount of adsorbed coupling agent on the fiber surface is not contributing in a proportional manner to either the IFSS or the tensile strength as shown by the micromechanical test results shown in Table 4. These results suggest that, when the adsorbed molecules have a low-molecular weight, the anchored chains, upon a mechanical solicitation act more efficiently than the molecular aggregates, which prevail at concentrations above the critical concentration, in this case is at approximately 0.005%. Furthermore, despite the efficiency of the fiber-surface modification, processing parameters such as initial fiber length, stresses during processing and time are not being considered in this analysis.

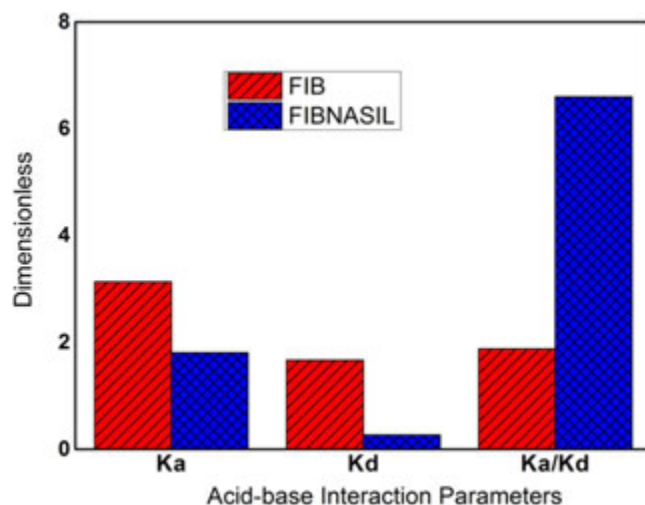


Fig. 10 K_a and K_d values for untreated (FIB) and silane treated (FIBNASIL) henequen fibers. Based on Valadez-Gonzalez, A., Moreno-Chulim, M.V., Herrera-Franco, P.J., 2009. Modification of the fiber surface for the optimization of mechanical properties in natural-fiber reinforced polymers. *International Journal of Materials and Product Technology* 36, 417–430. doi:10.1504/IJMPT.2009.027846.

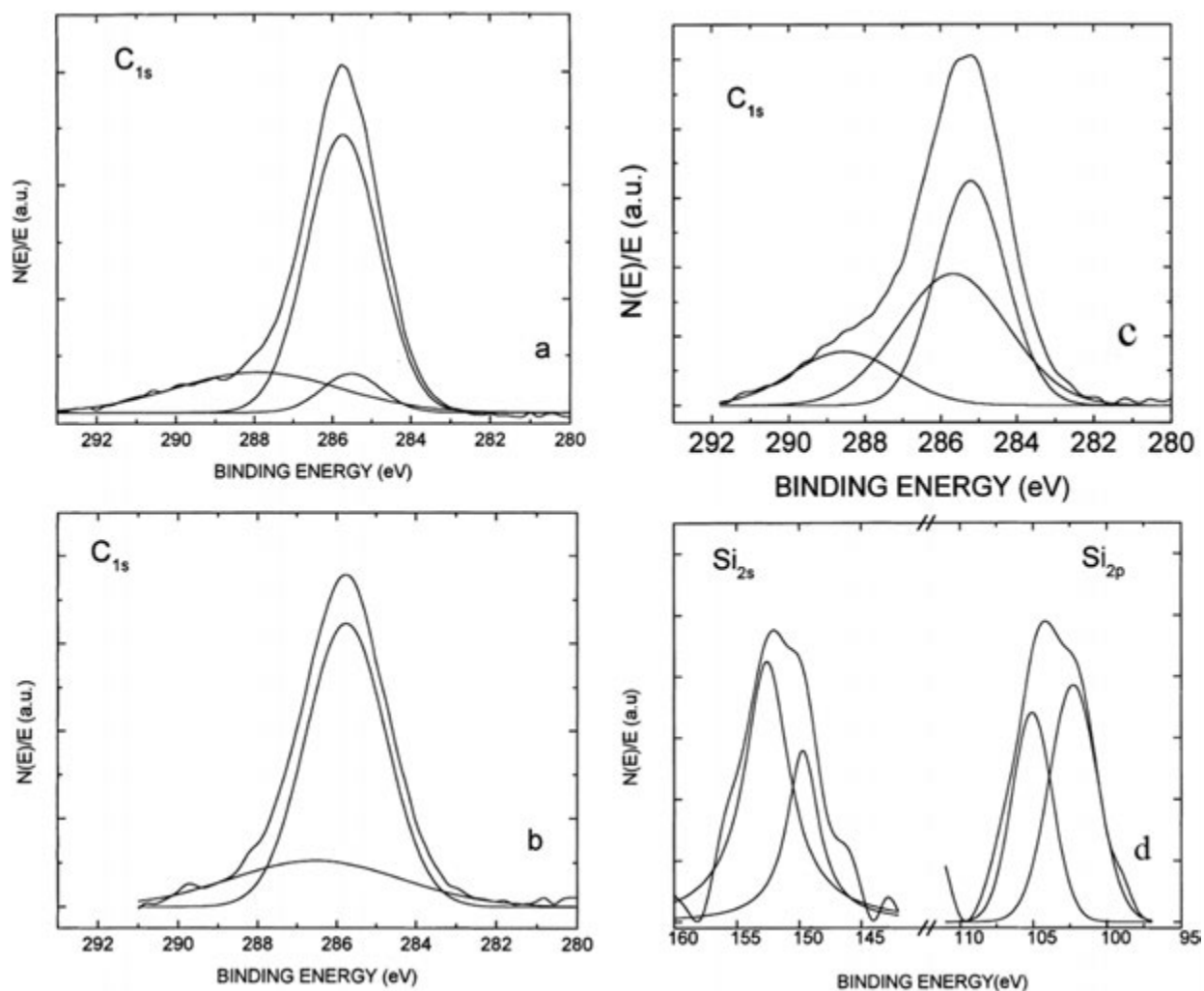


Fig. 11 XPS spectra of carbon peaks (C_{1s}) for (a) FIB; (b) FIBNA; (c) FIBNASIL and (d) silicon peaks (Si_{2s} and Si_{2p}) for FIBNASIL. Reprinted with permission from Valadez-Gonzalez, A., Cervantes-Uc, J.M., Olayo, R., Herrera-Franco, P.J., 1999b. Chemical modification of henequén fibers with an organosilane coupling agent. *Composites Part B: Engineering* 30, 321–331. doi:10.1016/S1359-8368(98)00055-9.

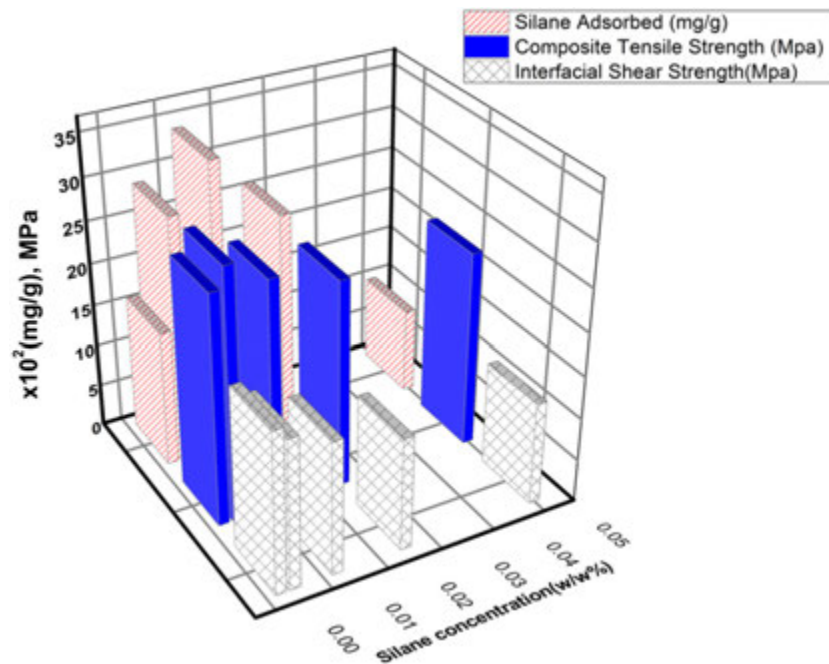


Fig. 12 Tensile and Iosipescu shear strengths of composite laminate as a function of the absorbed silane coupling agent on the fiber surface.

Table 4 Interfacial shear strength from the micromechanical tests

Henequen fiber surface treatment	IFSS (Pull-out) (MPa)	IFSS (SFFT) (MPa)	Shear strength iosipescu (MPa)
FIB	2.2 ± 0.81	4.39 ± 0.69	9.00 ± 0.69
FIBNASIL	5.2 ± 2.18	15.7 ± 2.98	14.4 ± 0.61

Conclusions

Natural fiber composites offer immense opportunities as alternative materials in several industrial applications. In addition to the attractive physical and mechanical properties, they are relevant to developing countries in view of their low cost, and savings in energy. The issue of the incompatibility between the naturally hydrophilic plant fibers and the hydrophobic polymer matrix has been amply studied and several solutions are available in the technical literature. The IFSS between natural fibers and different matrices, polymeric and cementitious matrices has been improved by morphological and chemical modifications of the fiber surface. Alkaline treatment increases surface roughness of the fiber and a better mechanical interlocking and also a larger amount of cellulose is exposed on the fiber surface and the number of possible reaction sites. The use of a silane coupling agent changed the surface physicochemical properties were measured. It was also found that the formation of polysiloxanes, inhibit the adsorption of the silane onto the henequen fiber surface. This fact is particularly important since at highly concentrated silane solutions there is no additional silane adsorption onto the fibers. From the XPS surface studies of the fibers, the presence of the silane group deposited on the surface (FIBNASIL) fibers was confirmed. The analysis of the emission spectra shows evidence of a chemical reaction between the hydrolyzed silane and the henequen fibers, at least as observed from the results for FIBNASIL. The fiber matrix adhesion was studied using micro mechanical techniques and the IFSS relationships developed for circular fibers were modified to incorporate the natural fiber perimeter instead of an equivalent fiber diameter. The use of the fiber critical aspect ratio (l/d) in the Weibull analysis resulted in higher values for IFSS but the trends were the same as those obtained from the use of the fiber cross-section perimeter for both the fragmentation test and the pull-out test. The results obtained from the single fiber fragmentation test seem to be in better agreement with the effective mechanical properties measured for the laminated material. The level of fiber-matrix adhesion was further enhanced by the presence of a silane-coupling agent. The fiber-surface silanization resulted in better interfacial load transfer efficiency.

It has been shown that the IFSS can be optimized in terms of the absorption of the coupling agent on the fiber surface as proved with both, single-fiber micromechanical techniques and laminate standard mechanical tests. However, the improvement observed on single-fiber-matrix adhesion is not observed in the same proportion in mechanical properties of the composite material laminate.

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Multiscale Structure of Plant Fibers

Christophe Baley and Alain Bourmaud, South Brittany University, Lorient, France

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Introduction

Introduction to the Reinforcement of Polymers With Plant Fibers

This article is dedicated to the multi-scale structure of plant fibers used for polymer reinforcement; these composite materials are called biocomposites. It is designed and intended for material specialists and not biologists, which explains the selections and simplifications.

Pushed by legislation and the increase in the environmental awareness of the public, the current development and use of biocomposites reduces environmental impacts and also valorizes and promotes local resources. In this context, a wide range of plant cells can be used as reinforcement and not only those used for textile applications. Moreover, in addition to their life cycle analyses, which are often much more favorable than those of synthetic fibers, plant fibers have intrinsic specificities that can be transformed into technological and scientific assets.

Biocomposites have special features, but they are first and foremost composite materials. Compared to industrially produced synthetic fibers, the use of renewable resources leads to new constraints, especially regarding fiber transformation or the evolution of their properties during the composite process. For an optimization of their use (to select and obtain reproducible properties), it is necessary to have a sound knowledge of this type of cell wall and all the pertinent parameters (sampling area in plants, multiscale structure, geometry, biochemical composition, geometrical and mechanical properties, parameters influencing their properties...).

What is a Fiber?

A plant has a hierarchical cellular structure with porous and dense cells that provide nutrient transport and/or mechanical support, a cell itself can have several specific functions within the plant. Thus, the plant develops the cells it needs to live and grow. “Fiber” is a term used by many researchers using natural fibers in composites. However, it has a strict botanical meaning, which describes a single elongated, thick-walled, plant cell. Among these fibers, those with a structural and supporting function generally have the best mechanical properties; however, many plant cells can be used to reinforce polymers. The diversity of plants and cells gives the opportunity to benefit from a wide range of possible reinforcements, but also to target and design specific use properties, well adapted to both the fiber and the application use considered.

Chapter Presentation

The aim of this article is to present a set of data to illustrate the multi-scale structure of plant fibers, potentially usable for composite reinforcement. It does not pretend to be complete, but aims to illustrate the diversity of plant fibers. Regardless of the considered species, and conversely to synthetic ones, there are no continuous fibers in the plant world; they all occur in a discontinuous form. Nevertheless, according to their properties and geometry, they can be successfully used for textile manufacturing (elementary fibers called “long” or assembly of fibers in a bundle) and papermaking (very short cells). Here, we focus on three plant fibers species or families: wood, flax and bamboo, notable by their structure; their differences provide a useful and interesting study case. A large section of this article is dedicated to the description of wood fibers. Behind this very generic term, a wide range of cells exist, whose properties change according to the environmental conditions, the considered species or varieties, and also to the function of the cells inside the plant.

Classification and Average Properties of Plant Fibers

Origin of the Main Plant Fibers Used as Reinforcement for Composite Materials

In the past, people have used plant fibers for making ropes, assemblies (ligatures) and textiles. They were locally collected in the natural environment, then the domestication and development of agriculture made it possible to multiply their uses. The main textile plant fibers come from cotton (mainly), flax, hemp, ramie, jute, kapok, agave (sisal, henequen), bananas (abaca or Manila hemp...) and palm (raffia, coir, ...) (Reis *et al.*, 2006). Nevertheless, not all textile fibers have good enough properties to obtain an optimum reinforcement if used for composite applications. In addition, the reinforcement mechanisms of a yarn (or a woven textile) are different from those of a composite material. For composites, two parameters are particularly important for the choice of the reinforcement (fibers): its aspect ratio (length divided by diameter) and its mechanical properties. From plants, fibers can have different botanical origins (Fig. 1).

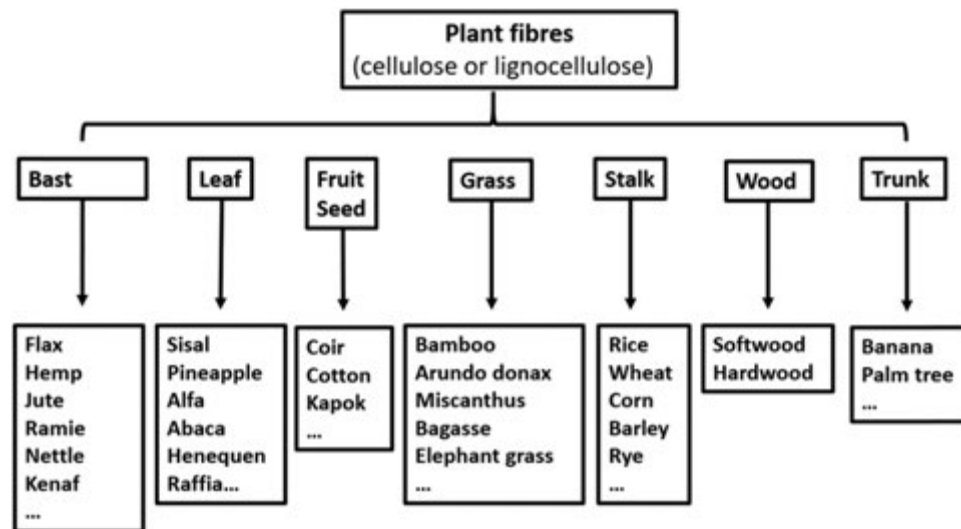


Fig. 1 Classification of plant fibers.

Plant fibers can come from bast fiber plants (flax, hemp...), leaves (sisal...), fruits (coir...), grass (Arundo donax...), straw (wheat...), wood (coniferous, deciduous) or pseudo-trunks (banana...). Usually, the plant fibers coming from the stems and leaves have the highest mechanical properties, mainly due to their mechanical role within the plant.

Comparison of the Geometrical Characteristics of the Main Plant Fibers Used as Reinforcement

In order to choose which plant fibers to use to reinforce composite materials, a range of intrinsic or exogenous criteria must be taken into account. The main ones to consider are:

- (1) Their geometry (Table 1),
- (2) Their biochemical composition (Table 1),
- (3) Their mechanical properties (Table 2),
- (4) Their cost which is a function of the main markets,
- (5) The volume of fiber available with reproducible properties,
- (6) The possibility of using them without specific adaptation of the composite manufacturing process,
- (7) The knowledge of external impacts being taken into account at all stages (production, retting, mechanical or other treatments, storage, transport, etc.).

Table 1 shows the average geometrical characteristics found in the literature, the microfibrillar Angle (MFA) and biochemical composition of different plant fibers used for polymer reinforcement. Fibers are assembled in plants in the shape of bundles whose geometry is very different from that of elementary fibers. Most of the elementary fibers have a short length (a few mm) except the fibers of flax, hemp, ramie, cotton and kapok (a few tens of mm), mainly due to their intrusive elongation mode during the growth of the plant.

Chemical Composition

Plant fibers can be considered as composite structures of oriented cellulose fibrils embedded in a matrix of more or less rigid structures formed by the hemicelluloses, pectin and lignin polymers (Table 1). The principal characteristics for fiber cells are determined by the amount and the distribution of the chemical constituents in various layers of the cell wall (this subject will be illustrated and discussed further, in the rest of the article).

The main constituents (Table 1) of the plant walls are:

- (1) Cellulose: The main stress-bearing compound of the plant cell wall is the crystalline cellulose microfibril, which typically consists of 10 nm wide crystallites of long and straight closely packed polymeric chains of exclusively β -(1–4)-linked D-glucopyranose units (van Dam and Gorshkova, 2003).
- (2) Noncellulosic cell wall polysaccharides (hemicelluloses, pectin, lignin): Though cellulose constitutes the major part of the wall, a number of other polysaccharides occur and are fundamental for fiber properties. Quantification of cell wall polysaccharides is complex due to various types of bonds between the individual polysaccharides and/or other components.

For a more in-depth look at the composition of plant cells, the reader can consult the following references (Evert, 2006; Evert *et al.*, 2012).

Table 1 Physical properties, microfibrillar Angle (MFA) and biochemical composition of different plant fibers that can be used to reinforce a polymer

	Bundle	Elementary fiber			Biochemical composition					
Name	Length (cm)	Length (mm)	Diameter (μm)	Aspect ratio	MFA (°)	Cellulose (%)	Pectins (%)	Hemicelluloses (%)	Lignin (%)	References
Abaca	100–200	3–12	12–36	312	5–17	62	5	13	11	(van Dam and Gorshkova, 2003)
Alfa		2–5	5–10	466		44–48.	1	26–38	15–23	(Bourmaud <i>et al.</i> , 2018)
Arundo donax		1.2	15–17	75		29	13	32	21	(El-Abbassi <i>et al.</i> , 2020)
Bamboo	20–34	0,5 – 5	5–20	220	8–11	36–55	<1	12–17	20–29	(Bourmaud <i>et al.</i> , 2018)
Coir	5–20	0,3–1	12–24	36	30–49	33	5	13	33	(Shatalov and Pereira, 2002)
Cotton (trichomes)	–	20–60	12–25	2162	20–30	90	–	6	<1	(Ren <i>et al.</i> , 2014)
Flax	60–110	13–60	12–30	1738	10	75	3	15	<1	(Yu <i>et al.</i> , 2011)
Hardwood		0.7–1.6	20–30	50	5–40	31–64	0.1–8	24–40	14–34	(van Dam and Gorshkova, 2003)
Hemp	100–300	5–55	16–50	909	11	70	2	16	3	(Bourmaud <i>et al.</i> , 2018)
Jute	15–36	0,8–7	5–25	260	7–12	62	1	22	13	(van Dam and Gorshkova, 2003)
Kapok	–	10–20	10–30	750	5	43	4	23	15	(Bourmaud <i>et al.</i> , 2018)
Kenaf	90–180	1,5–11	14–33	266	9–15	55	4	13	12	(van Dam and Gorshkova, 2003)
Miscanthus		0.97	14.2	68		43.7			28.5	(Ververis <i>et al.</i> , 2004)
Pineapple leaves		3–9	6–80	139	8–15	70–85	1	–	12–15	(Alwani <i>et al.</i> , 2014)
Ramie	>150	50–200	15–80	2631	7,5	76	2	15	1	(Jawaid and Abdul Khalil, 2011)
Rice		0.4–1.2	8–15.5	68		34	11	23	17	(Satyanarayana <i>et al.</i> , 2007)
Sisal	60–100	0,8–8	10–40	176	20	73	1	13	11	(van Dam and Gorshkova, 2003)
Softwood		2.7 – 4.6	32–43	100	7–40	30–60	0.2–8.5	20–30	21–37	(Bourmaud <i>et al.</i> , 2018)
Wheat straw		0.4–3.2	8–34	86		30	5	50	15	(Jawaid and Abdul Khalil, 2011)
										(Tsoumis, 1991)
										(Eder and Burgert, 2010)
										(Juliana <i>et al.</i> , 2018)
										(Khalil <i>et al.</i> , 2012)
										(Ilvessalo-Pfäffli, 1995)
										(Sain and Panthapulakkal, 2006)

Table 2 Bibliographic synthesis of mechanical properties of cellulosic fibers elements (elementary fibers or bundles), with E_{FL} : longitudinal Young's modulus (GPa), A_{FLU} ultimate elongation (%) and σ_{FLU} strength to break (MPa) and density

Name	E_{FL} (GPa)	A_{FLU} (%)	σ_{FLU} (MPa)	Density	References
Abaca	8–20	2–8	400–980	1.35	(Cai <i>et al.</i> , 2015)
Alfa	18–22	1,4–2,4	229–474	1,4	(Mamun <i>et al.</i> , 2015)
Aroundo donax	9.4	3.24	248		(El-Abbassi <i>et al.</i> , 2020)
Bamboo	32.0–43.7	3.8–5.8	1200–1610	1.41	(Helaili and Chafra, 2014)
Cair	4–6	15–40	131–175	1.15	(Khalidi <i>et al.</i> , 2016)
Cotton	5.5–13	3–10	287–800	1.5–1.6	(Scalici <i>et al.</i> , 2016)
Flax	38–75	1,7–3	600–1400	1.5	(Ren <i>et al.</i> , 2014)
Hardwood	30.7		866	0.3–0.88	(Yu <i>et al.</i> , 2011)
Hemp	23.5–90	1–3.5	270–900	1.45	(Geethamma <i>et al.</i> , 1998)
Jute	20–30	1.5–1.8	280–773	1.44	(Pickering <i>et al.</i> , 2016)
Kenaf	21 – 80	1.6	284–800	1.40	(Bledzki and Gassan, 1999)
Nettle	87	2.11	1594		(Bourmaud <i>et al.</i> , 2018)
Pineapple leaves	4.4–36	2 – 2.8	126 – 748	1.4	(Baley and Bourmaud, 2014)
Ramie	24.5–65	1.6–3.8	331–1250	1.51	(Eder and Burgert, 2010)
Rice	3.3–12.5	3.2–4.6	152–200		(Gurunathan <i>et al.</i> , 2015)
Sisal	9–21	3–7	350–700	1.45	(Bourmaud and Baley, 2009)
Softwood	16–26		386–930	0.3–0.59	(Duval <i>et al.</i> , 2011)
Wheat straw	3.7–4.8		59–140		(Marrot <i>et al.</i> , 2013)

Mechanical Properties

Table 2 shows the mechanical tensile properties of various fibers from a wide range of botanical origins. Since some fibers are very short, it is highly probable that the tensile tests were carried out on bundles and not on elementary fibers. Given the natural origin of these fibers, scattering properties can be observed. For a certain type of fiber, this scattering can be explained by several parameters such as the origin, the variety (genetic heritage), the growing and harvesting conditions of the fibers (plant maturity), retting and the treatments they have undergone.

Therefore, one should not quickly draw conclusions over the superiority or lack of usefulness of any particular variety. Moreover, in the literature, the total characterization conditions (equipment, sample geometry, tensile speed, temperature, humidity, characterization of a single fiber or a bundle, result analyses ...) are not always identical. The standardization of the definition of tensile characterization conditions of flax fibers is currently taking place (AFNOR NTT 25–501–1, 2015; AFNOR NFT 25–501–2, 2015; AFNOR NF T 25–501–3, 2015).

Table 2 shows a bibliographic synthesis of the mechanical properties of cellulosic fibers elements (elementary fibers or bundles), with E_{FL} : longitudinal Young's modulus (GPa), A_{FLU} ultimate elongation (%) and σ_{FLU} strength to break (MPa) and density. (Cai *et al.*, 2015; Mamun *et al.*, 2015; El-Abbassi *et al.*, 2020; Helaili and Chafra, 2014; Khalidi *et al.*, 2016; Scalici *et al.*, 2016; Ren *et al.*, 2014; Yu *et al.*, 2011; Geethamma *et al.*, 1998; Pickering *et al.*, 2016; Bledzki and Gassan, 1999; Bourmaud *et al.*, 2018; Baley and Bourmaud, 2014; Eder and Burgert, 2010; Gurunathan *et al.*, 2015; Bourmaud and Baley, 2009; Duval *et al.*, 2011; Marrot *et al.*, 2013;

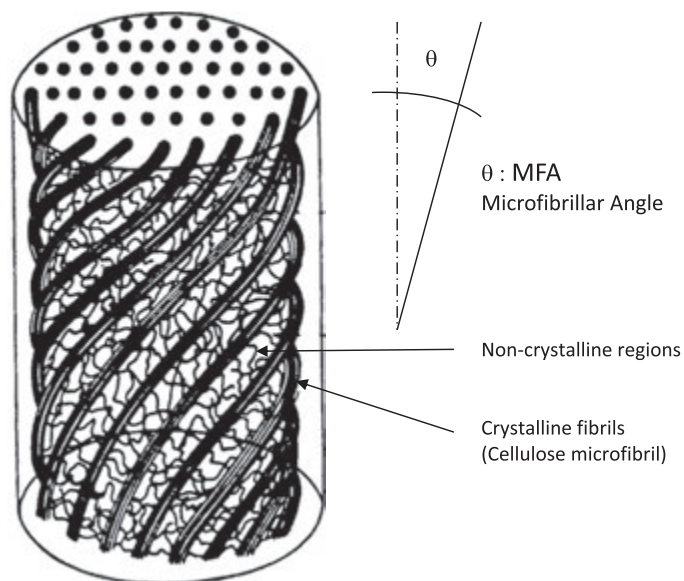


Fig. 2 Schematic drawing of the structure of a plant fiber with a helical arrangement of cellulose fibrils. Reproduced from Hearle, J., 1963. The fine structure of fibers and crystalline polymers. III. Interpretation of the mechanical properties of fibers. *J. Appl. Polym. Sci.* 7, 1207–1223. Available at: <https://doi.org/10.1002/app.1963.070070403>.

Roe and Ansell, 1985; Tanguy *et al.*, 2016; Lee *et al.*, 2009; Akil *et al.*, 2011; Saba *et al.*, 2015; Bodros and Baley, 2008; Jawaid and Abdul Khalil, 2011; Arib *et al.*, 2006; Satyanarayana *et al.*, 2007; Angelini *et al.*, 2000; Du *et al.*, 2015; Fuqua *et al.*, 2012; Zhao *et al.*, 2011; Li *et al.*, 2000; de Andrade Silva *et al.*, 2008; Eder and Burgert, 2010; Gurunathan *et al.*, 2015; Fuqua *et al.*, 2012; Sain and Panthapulakkal, 2006; Hornsby *et al.*, 1997a; Hornsby *et al.*, 1997b).

Simplified Structure of a Plant Fiber

In the first approach, a plant fiber is comparable to a composite material reinforced by cellulose fibrils (Fig. 2) (Hearle, 1963). The matrix is mainly composed of hemicelluloses, pectin and lignin. The cellulose fibrils are helically oriented at an angle called the microfibrillar angle (MFA) (See Fig. 2 and Table 1). The microfibrillar angle is the angle between the fibrils and the longitudinal axis.

Usually, in a composite material, the fiber volume fraction and the fiber orientation determine the elastic and break properties. Similarly, in a plant fiber, the physical properties of natural fibers are mainly governed by the biochemical composition, nanostructure, percentage of cellulose and its crystallization degree, the microfibrillar angle (Bledzki and Gassan, 1999) and the properties of the polysaccharides ensuring the transfer of loads between the fibrils. To simplify, for a given percentage of cellulose, with a small microfibrillar angle (Table 1) the fibers have a high stiffness and tensile strength. Otherwise, with a large microfibrillar angle, the strain at break is greater. The impact of the cellulose content and MFA on the cell wall stiffness has been modeled on wood (Jäger *et al.*, 2011). These calculations make it possible to predict the expected performance according to the ultrastructure of the fibers.

Tables 1 and 2 confirm this, showing that the most suitable fibers to reinforce a polymer (flax, hemp, nettle and ramie) have a high aspect ratio, a high cellulose content and a low microfibrillar angle. A significant length will be an advantage for the preforms manufacturing. Nevertheless, elementary fibers with a short length are not to be excluded because bundles can be used. In addition, many composites processed by extrusion or injection are reinforced with short fibers (wood fibers for example) of a few mm in length.

In reality a fiber is a multi-layered cell wall reinforced by cellulose fibrils, but the biochemical composition, thickness and orientation change according to the considered layer. Fig. 3 shows organizational models and associated vocabulary.

The next section is devoted to wood fibers whose multi-scale structure is often taken as a reference for plant fibers.

Wood Fiber, A Reference

Overall Presentation of Different Wood Fibers

In this context, wood fibers are taken as an example and model to present the multi-layered structure of plant fibers. Trees are generally classified into two families: softwood and hardwood. The trunk of each family contains different cells each one performing specific functions. In the softwoods, the three functions (storage, support and conduction) are performed by two types of cells, whereas in the hardwoods each function is performed by a single cell type (Table 3).

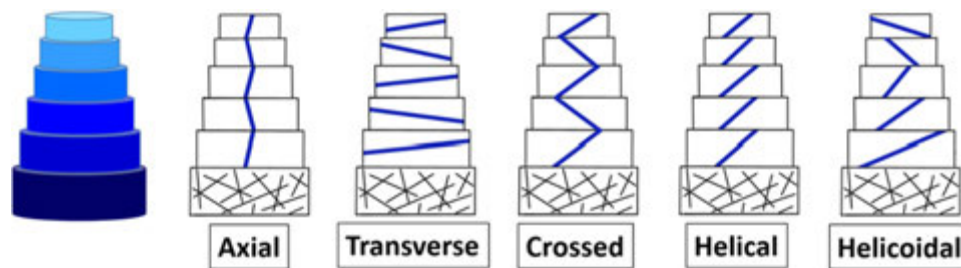


Fig. 3 Basic wall textures: axial, transverse, crossed, helical, helicoidal and random. Successive wall lamellae have been peeled off from top to bottom.

Table 3 Functions and typical cross section of the various types of cells found in softwoods and hardwoods

<i>Cells</i>	<i>Softwood</i>	<i>Hardwood</i>	<i>Function</i>	<i>Typical cross section wall thickness</i>
Parenchyma	X	X	Storage	
Tracheids	X	X	Support, Conduction	
Fibers		X	Support	
Vessels (pores)		X	Conduction	

Note: Adapted from Dinwoodie, J.M., 2004. Timber: Its Nature and Behaviour, second ed. Taylor & Francis Group.

These different types of cells are described in the books dedicated to wood but also in those focusing on pulp and paper (Ilvessalo-Pfäffli, 1995; Biermann, 1995). As for paper, using wood fibers as a composite reinforcement means using different types of cells. Table 3 shows the functions and typical cross sections of the various types of cells found in softwoods and hardwoods (Dinwoodie, 2004). In the softwood two types of cells can be observed. Those present in greater number are known as tracheids, some 2–4 mm in length with an aspect ratio (L/D) of about 100:1. These cells, which lie vertically in the tree trunk, are responsible for both the supporting and conducting roles. The small block-like cells some $200 \times 30 \mu\text{m}$ in size, known as parenchyma, are mostly located in the rays and are responsible for the storage of food material (Dinwoodie, 2004).

In contrast, in the hardwoods, four types of cells are present, albeit that one, the tracheid, is present in small amounts. The role of storage is again primarily taken by the parenchyma, which can be present horizontally in the form of a ray, or vertically, either scattered or in distinct zones. Support is ensured by long thin cells with very tapered ends, called fibers; these are usually about 1–2 mm in length with an aspect ratio of about 100:1. Conduction is carried out through cells whose end walls have been dissolved away, either completely or in part. These cells, known as vessels or pores, are usually short (0.2–1.2 mm) and relatively wide (up to 0.5 mm) and when situated above one another form an efficient conducting tube (Table 3).

Description of a Tracheid

As an example, Fig. 4 shows a softwood tracheid, illustrating a laminate organization (Fig. 4). Softwood tracheids are approximately $30 \mu\text{m}$ wide and contain a number of distinct features including thin cell walls, pits and a distinct transition between

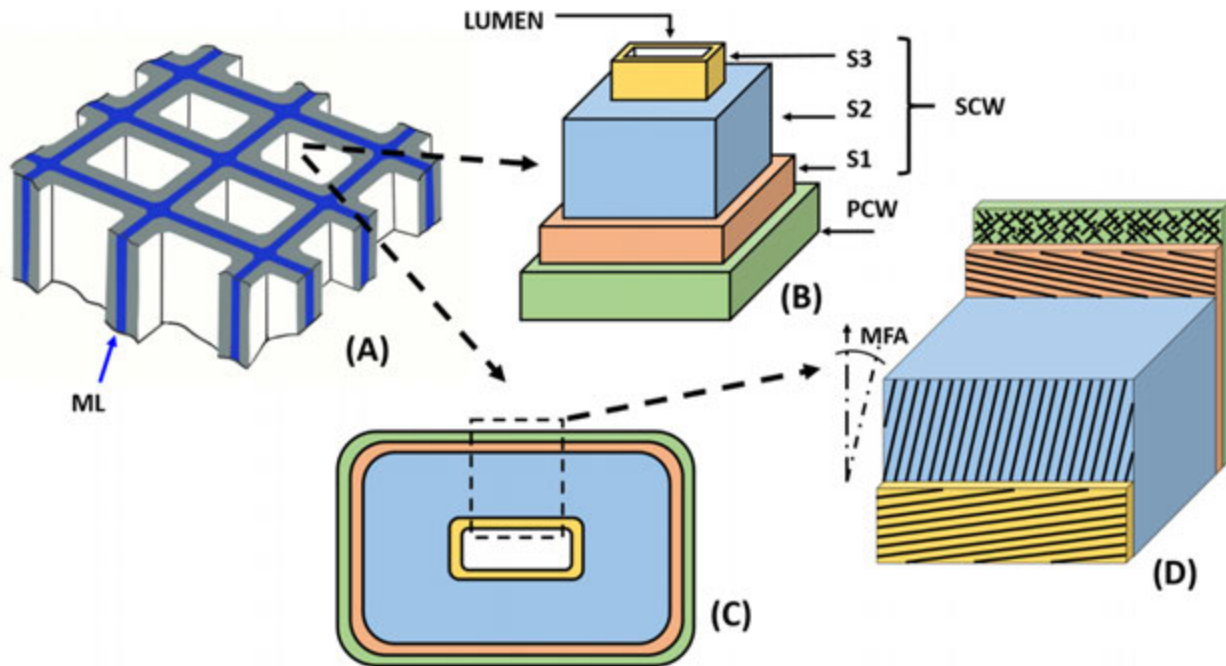


Fig. 4 Cellular structure of earlywood in softwoods and structure of softwood tracheids showing primary and secondary cell walls and orientation of cellulose microfibrils (black lines). ML = Middle Lamella. PCW: Primary Cell Wall, SCW: Secondary Cell Wall (with layers S1, S2, and S3).

Table 4 Chemical composition of timber

Component	Mass		Polymeric state	Molecular derivatives	Function
	Softwood (%)	Hardwood (%)			
Cellulose	42 ± 2	45 ± 2	Crystalline, Highly oriented, Large linear molecule	Glucose	Reinforcement
Hemicelluloses	47 ± 2	30 ± 5	Semicrystalline, Smaller molecule	Galactose Mannose Xylose Phenylpropane	Matrix
Lignin	28 ± 3	20 ± 4	Amorphous, Large 3-D molecule		
Extractives	3 ± 2	5 ± 4	Principally compounds soluble in organic solvents	Terpenes, Polyphenols, Stillbenoids	Extraneous

Note: Adapted from Dinwoodie, J.M., 2004. Timber: Its Nature and Behaviour, second ed. Taylor & Francis Group.

earlywood and latewood. Some cells include internal helical thickening within the cell cavity which prevents the collapse of light weight thin cell walls under high suction during transpiration (Gibson, 2012; Ansell, 2015).

All tracheids contain both a primary and secondary wall, with the secondary wall being split into three separate layers: S1, S2 and S3 (Fig. 4). The secondary cell wall is added after the initial formation of the cell. Between them, the tracheids are bonded together by middle lamellae. The microfibrillar angle shown in Table 1 is that of the S2 layer, S2 is the main layer in a fiber. The hollow cavity inside the fiber is called the lumen.

The thicknesses of the layers are typically 0.1–0.3 μm, 1–5 μm and 0.1 μm for the S1, S2 and S3 and primary layer, respectively (Mark, 1967). The middle lamella (ML), consisting mainly of lignin (amorphous oxyphenyl propane units), connects adjacent cell walls as well as the lower content of polysaccharidic pectin (Whiting and Goring, 1982).

The overall chemical composition of a wood fiber is given in Table 1. In addition, Table 4 gives details on the nature and biochemical ultrastructure of the components. (adapted from (Dinwoodie, 2004)). Here, the proportions are generally given for timber; slight variations in these can occur according to the considered species. The nature of the constituents of each family is described. Within the different cell wall layers, cellulose exists as a system of fibrils of 3–4 nm in diameter aggregated in larger structural units. The cellulose microfibrils, ensuring the reinforcement of the cell walls, are helically wound at different angles in the various layers of the cell wall.

Table 5 Chemical composition of the cell wall of Scots pine

	<i>Middle Lamella + Primary cell wall (PCW)</i>	<i>Secondary cell wall (SCW)</i>			<i>Total Mass (%)</i>
		<i>S1</i>	<i>S2</i>	<i>S3</i>	
Cellulose (%)	0.7	6.1	32.7	0.8	40.3
Hemicelluloses (%)	1.4	3.7	18.4	5.2	28.7
Lignin (%)	8.4	10.5	9.1	–	28.0
Total / Total mass (%)	11	21	62	6	

Note: Adapted from Rowell, R., 2013. Handbook of Wood Chemistry and Wood Composites, second ed. CRC Press.

Table 6 Microfibrillar orientation and percentage thickness of the cell wall layers in spruce timber (*Picea abies*)

	<i>Primary cell wall</i>	<i>Secondary cell wall</i>		
		<i>S1</i>	<i>S2</i>	<i>S3</i>
Approximate Thickness (%)	3	10	85	2
MFA (°)	Random	50 – 70	10–30	60–90

Note: Adapted from Dinwoodie, J.M., 2004. Timber: Its Nature and Behaviour, second ed. Taylor & Francis Group.

Specifically, the constituent proportion is not the same in all layers (Schwarze, 2007). The content of cell wall components depends on the tree species and on the tissue origin. Softwoods are different from hardwoods, heartwood from sapwood and latewood from springwood.

Table 5 shows the distribution of components across the cell wall of Scots pine. For each layer, the fraction of each component is expressed as a percentage of the total weight. The middle lamella and primary wall are mainly composed of lignin (80%) with lesser amounts of hemicelluloses (13%) and even less cellulose (7%). The S1 layer is composed of 52% lignin, 30.0% cellulose, and 18% hemicelluloses. The S2 layer is composed of 15% lignin, 54% cellulose, and 31% hemicelluloses. The S3 layer has little or no lignin, 13% cellulose, and 87% hemicelluloses. The xylan content is lowest in the S2 layer and higher in the S1 and S3 layers. The concentration of galactoglucomannan is higher in the S2 than the S1 or S3 layers. On a percentage basis, the middle lamella and primary wall contain the highest concentration of lignin but there is more lignin in the S2 because it is a much thicker layer as compared to the middle lamella and primary wall. The lignin in the S2 layer is evenly distributed throughout the layer (Rowell, 2013).

The angle of the cellulose microfibrils in the various cell wall layers, in relation to the fiber axis is known as the microfibrillar angle (MFA). As an illustration, Table 6 shows the microfibrillar orientation and relative thickness of each cell wall layer in spruce timber (*Picea abies*). The S2 layer, due to its thickness and organization is the most structural one.

Transition Zone

Between each layer, the variation in the orientation of the cellulose fibrils is not abrupt but progressive. There are transition zones (between S1 and S2 and between S2 and S3) observable by TEM (Transmission Electron Microscopy) (Roland *et al.*, 1995; Wang *et al.*, 2013; Reza *et al.*, 2014, 2015). In these transition zones, microfibrils stay in parallel planes and change their orientation progressively. This topic is detailed in Section “Flax Fibers, Multi-Scale Structure and Sources for Bioinspiration”, dedicated to flax fibers.

In-Depth Description of the S₂ Layer, From the Global Architecture to Cellulose Fibrils

The characterization of plant cell walls is not easy. Typical approaches to study the chemical composition, structure and architecture of plant cell walls include microscopy, spectroscopy and scattering techniques such as X-Ray diffraction.

At the molecular level, Nuclear Magnetic Resonance (NMR), Fourier Transform Infrared Spectroscopy (FT-IR) and Raman spectroscopy (Agarwal, 2014) can provide information regarding the overall and localized chemical composition of plant cell walls, as well as the chain conformation and intra- and intermolecular bonds, such as hydrogen bonds between hydroxyl groups. The arrangement of the cellulose molecules into ordered domains or cellulose crystallites can be studied by means of diffraction techniques. In particular, X-ray diffraction (XRD) has been widely used to resolve the crystalline configuration of cellulose microfibrils, i.e. to estimate the crystallite size and the cellulose crystalline fraction (Martínez-Sanz *et al.*, 2015). Nevertheless, crystallinity information obtained through NMR are often more precise as they specifically deal with cellulose and not a mix of components.

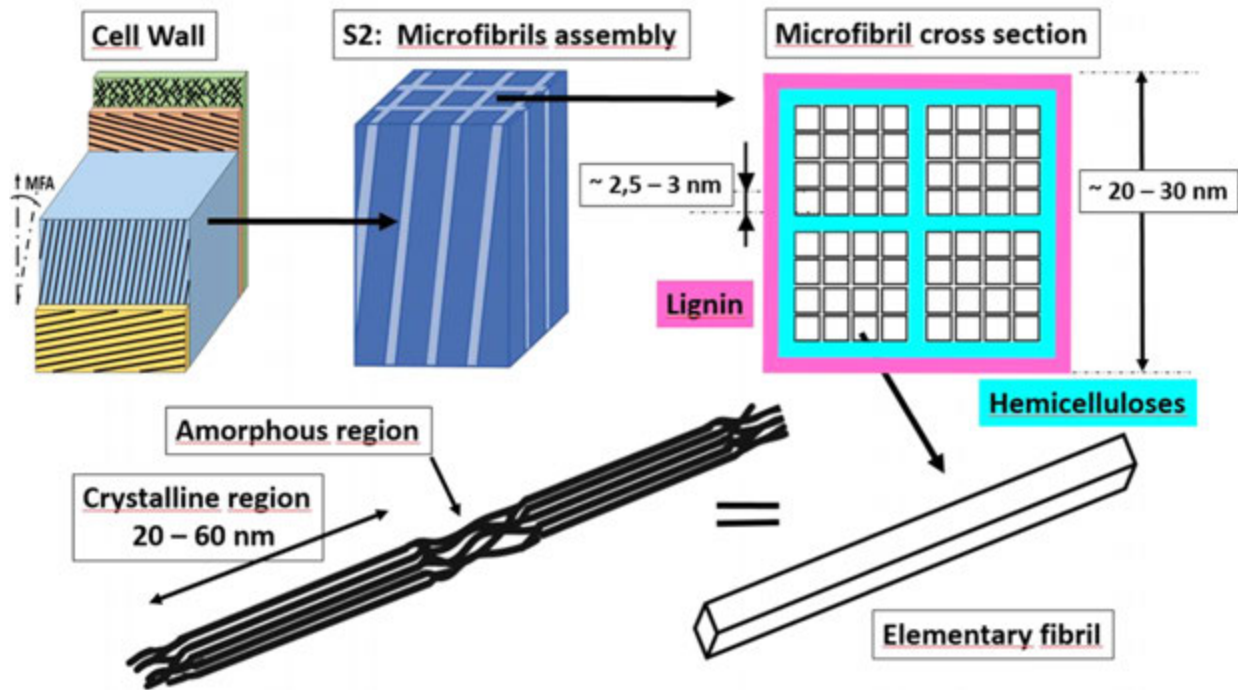


Fig. 5 Schematic drawing of layer S2 (secondary wall).

Covering both the nanostructural and microstructural levels, microscopy techniques have been the most frequently used approaches to investigate the structure of plant cell walls. Techniques such as scanning electron microscopy (SEM) and transmission electron microscopy (TEM) require specimen preparation and often involve drying the sample, which is likely to affect the structure of the material. As an alternative, environmental SEM (ESEM) and atomic force microscopy (AFM) have been used to analyze hydrated bacterial cellulose composites (Astley *et al.*, 2001) and cell walls (Davies and Harris, 2003; Song *et al.*, 2020).

The description of the S2 layer makes it possible to understand the origin of the properties of the fibers. The stiffness, strength and toughness of wood derive largely from the cellulose itself. The dominating S2 layer is composed of a series of helically wound cellulose microfibrils (CMFs), which are orientated under an acute angle (microfibril angle) towards the fiber axis. Several models exist to describe the arrangement of the chains within the microfibrils, but the models with folded chains are the most frequently used (Dufresne, 2013). Within the cell wall, the cellulose microfibrils reinforce an amorphous matrix consisting of lignin, hemicelluloses, proteins, extractive organic substances, and trace elements. The cellulose microfibrils and hemicelluloses are linked to each other by hydrogen bonds. On the other hand, the hemicelluloses are more strongly linked to lignin through covalent bonds, that is to say, the hemicellulose component is a compatibilizer between cellulose and lignin. Microfibrils have diameters and lengths depending on the botanical origin of the cellulose. The cellulose microfibrils generally have a diameter of 10–30 nm and are composed of 30–100 cellulose macromolecules in an extended chain conformation (Fig. 5).

An elementary fibril (diameter ~1–5 nm (Martínez-Sanz *et al.*, 2015)) consists of both crystalline and amorphous regions. Bundles of elementary fibrils further form microfibrils which are embedded in an amorphous matrix of lignin, hemicelluloses (and other compounds). Cellulose is a linear chain of ringed glucose monomers (10,000–15,000) linked together. Multiple cellulose chains are arranged to form cellulose microfibrils with regions that are disordered (amorphous regions) or highly ordered (crystalline regions). Crystalline microfibrils are built up of aligned molecules some 30–60 nm in length (Fig. 5).

Figs. 4 and 5 show a simplified wood tracheid model with several scales. They illustrate the hierarchical structure of a wood fiber. Attention must be paid to the fact that the vocabulary used in the literature is not always identical. Here, some selections have been made to facilitate the reader's understanding.

Comparison Between Primary and Secondary Cell Wall

It is interesting to compare the different cell walls. Here, we will focus on a simplified illustration of the underlying principles and concepts of primary and secondary cell walls. Generally speaking, both primary and secondary cell walls can be described by means of plant fiber composites consisting of a stiff fibrous phase made of cellulose fibrils composed of crystalline and amorphous regions, as well as a soft amorphous matrix made of various biopolymers with hydrogen bonding between the two phases. According to their specific functions, both cell-wall types can be distinguished by means of structural and chemical parameters. This applies to the cell-wall thickness, cellulose orientation, degree of crystallization, volume fractions of cell-wall components, composition of the matrix, chemical bonding patterns, and water content (Burgert and Keplinger, 2013). It is important to note

Table 7 Comparison of the general features of the primary and secondary cell walls

Feature	Primary cell walls	Secondary cell wall
Cell wall	Thin	Thick, multi-lamellar, fully differentiated structure
Polymer network	Flexible network, ongoing modification processes	Rigid network, interlocked status
Cellulose	Low content, reorientation possible, orientation more variable	High content, densely packed, strictly parallel orientation
Matrix	Predominately hemicelluloses, pectin, and structural proteins	Predominately hemicelluloses and lignin
Water interactions	Highly hydrophilic, hydrogel character	More hydrophobic when lignified

Note: Adapted from Burgert, I., Keplinger, T., 2013. Plant micro- and nanomechanics: Experimental techniques for plant cell-wall analysis. J. Exp. Bot. 64, 4635–4649. Available at: <https://doi.org/10.1093/jxb/ert255>.

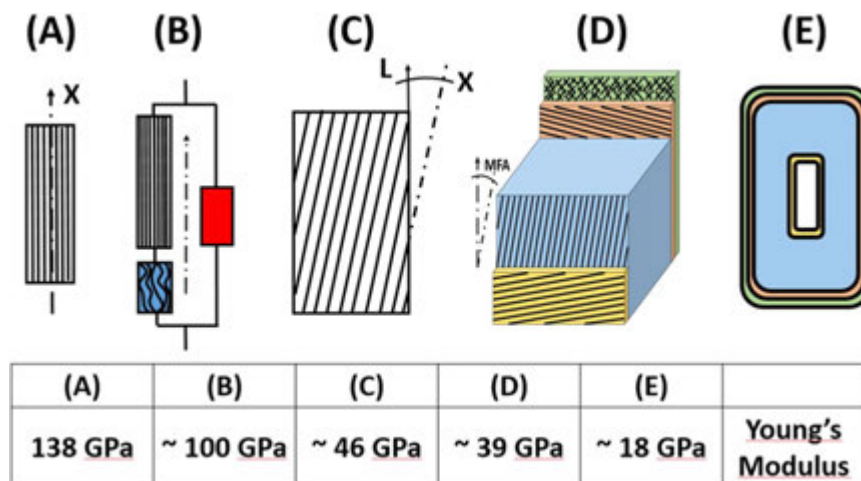


Fig. 6 Bottom-up description of the evolution of the Young's modulus from cellulose to wood fiber. Stiffness estimated by calculation (such as an anisotropic material) with the following assumptions: MFA = 15°, S2 = 85% of the total wall area, volume fraction of cell material = 50%.

that all these parameters also vary highly within primary and secondary cell walls. In [Table 7](#), the parameters most influencing the mechanical properties of primary and secondary cell walls are listed.

Relationship Between the Mechanical Properties of a Cellulose Fibril and Those of a Wood Fiber

The elastic modulus of the crystalline regions of cellulose polymorphs in the direction parallel to the chain axis was measured at 137 GPa by [Sakurada et al. \(1962\)](#) and 138 GPa by [Nishino et al. \(1995\)](#).

Overall, the mean modulus value for cellulose microfibrils is of the order of 100 GPa, while for cellulose crystal it is of the order of 138 GPa ([Dufresne, 2013](#)). By comparison, the lignin modulus is estimated to be 4 GPa and that of hemicelluloses 8 GPa ([Cousins, 1978](#)). However, the stiffness of hemicelluloses is strongly dependent on the moisture content. With 12% of absorbed water (approximate value if wood is stored at room temperature), the hemicelluloses modulus is only 2 GPa ([Salmen, 2004](#)). Thus, there is a large difference between the stiffness of the crystalline cellulose (the reinforcement) and the other constituents (the matrix).

The longitudinal modulus of a wood fiber is between 16 and 30 GPa ([Table 2](#)), which is significantly low compared to that of the crystalline region of an elementary fibril (138 GPa). To understand this gap and through a bottom-up approach, [Fig. 6](#) illustrates the progressive evolution of stiffness, according to the considered scale, i.e.:

- (1) [Fig. 6\(A\)](#): Crystalline cellulose (modulus of 138 GPa)
- (2) [Fig. 6\(B\)](#): The S2 layer organization: an assembly in parallel with the cellulose (series assembly of crystallized and amorphous regions with a stiffness of about 100 GPa) and the matrix (mainly hemicelluloses and lignin). The average percentage of cellulose is shown in [Tables 1, 4](#) and [5](#).
- (3) [Fig. 6\(C\)](#): In the S2 layer of the secondary wall, the fibrils have an orientation which differs from the longitudinal direction, which results in a decrease of the stiffness. Values of MFA are given in [Tables 1](#) and [6](#).
- (4) [Fig. 6\(D\)](#): The S2 layer holds the main the thickness, but other layers (S1 and S3 of the secondary cell wall and primary cell wall), having a lower stiffness co-exist. Although woods differ enormously in their density and mechanical properties, the properties of the single cell are, as a rough approximation, quite similar for all considered woods ([Gibson, 2012](#)). The cell wall laminate has a stiffness between 25 and 35 GPa ([Gibson, 2012](#)). This value depends on many parameters such as the

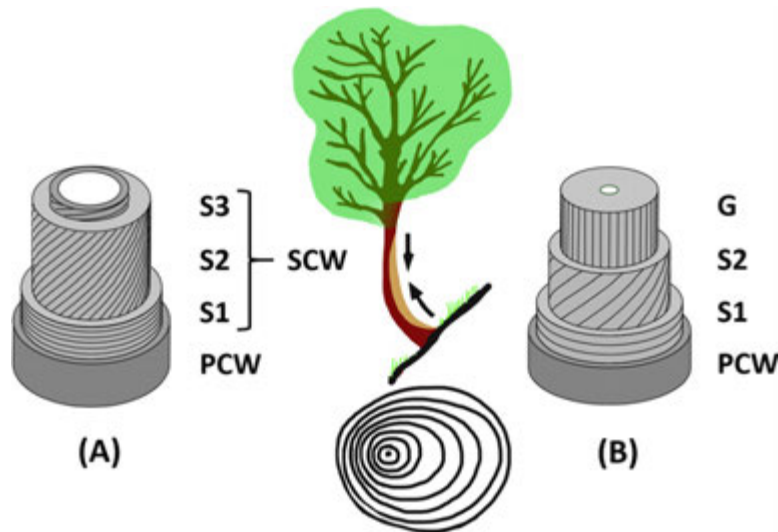


Fig. 7 Tree (Hardwood) that has straightened during growth. In other color, the part of the trunk containing tension wood is shown. Schematic models for the cell wall structure of fibers in normal wood (A) and tension wood (B) with a G-layer.

constituents, the microfibrillar angle (MFA), the moisture content, the method and characterization type, the volume of material tested, the maturity of the cells and the sampling area in the plant and the layer's thickness. Due to extraction issues and low fiber length, it is difficult to experimentally measure this stiffness directly. Nevertheless, measurements can be made, for example, by tensile tests on wood specimens taking into account the porosities (lumens) (Cave, 1969), by tensile tests on a tracheid (Gierlinger *et al.*, 2006) or by nanoindentation and AFM (local characterization in this case) (Eder *et al.*, 2013; Jäger *et al.*, 2011; Melelli *et al.*, 2020).

- (5) **Fig. 6(E)**: A fiber has a hollow cavity (lumen) which is necessary to take into account to determine the real section. Knowing the fiber density, the stiffness can be estimated. Indeed, the cellular structure of wood can be modeled, to first order, as a honeycomb with prismatic cells. The Young's modulus of wood along the grain varies linearly with the relative density (cell wall has a density close to 1500 kg/m³) (Gibson, 2012).

Wood Fiber Microstructure can Evolve to Adapt, e.g., Tension Wood

Reaction wood has been defined by the Nomenclature Committee of the International Association of Wood Anatomists (IAWA, 1964) as "Wood with more or less distinctive anatomical characters, formed typically in parts of leaning or crooked stems and in branches and tending to restore the original position, if this has been disturbed. It is divided into two types: tension wood in dicotyledons (hardwood) and compression wood in conifers (softwood)". Only tension wood will be presented in this section. Tension wood is: "Reaction wood formed typically on the upper sides of branches and on leaning or crooked stems of dicotyledonous trees and characterized anatomically by lack of cell wall lignification and often by the presence of an internal gelatinous layer in the fibers" (Gardiner *et al.*, 2014).

Tension wood fibers are longer than normal wood fibers and have been found to contain a lower proportion of lignin (Alm  ras and Clair, 2016). They are most commonly described as a normal fiber for which the S3 layer no longer exists and has been replaced by a gelatinous layer composed mainly of hydrated cellulose microfibrils oriented almost parallel to the long axis of the fiber, called S3 or the G-layer due to its gelatinous character (Fig. 7).

Fig. 7 shows an example of a tree that had been laid over by a landslide and then straightened as it grew. The function of the reaction wood (yellow area in Fig. 7) is to restore the displaced stem (trunk) to its normal vertical orientation. The percentage of cellulose changes in the presence of a G-layer from 39% to 45% to 50%–65% (Sj  str  m and Alen, 1999). Interestingly, the term G-layer is also used to describe the microstructure of a flax fiber due to the strong similarities between the ultrastructure and properties of flax and tension wood fibers (Gorshkova *et al.*, 2015). However, as described in the next section, mechanical properties of flax fibers are significantly higher.

Flax Fibers, Multi-Scale Structure and Sources for Bioinspiration

The aim of this section is to precisely describe the flax fiber microstructure and its specific construction during its growth.

Fiber Description

Fig. 8 illustrates the architecture of a mature flax fiber. This overall architecture is close to that of a wood fiber (**Fig. 4**). However, it has some fundamental differences, especially regarding the thicknesses of the S2 and S3 layers as well as the lumen size.

A flax fiber is made up of a thin primary wall surrounding a thick secondary wall (with three layers S1, S2 also called G-layer and S3 or Gn-layer, terms classically used for vegetal fibers (Rihouey *et al.*, 2017)), which is reinforced by cellulose microfibrils (analogous to a stacking of nanostructured plies), and a cavity in the middle which is called the lumen (**Fig. 8**). In a mature flax fiber, the lumen is estimated to represent 6.8% of the total fiber cross-section (Charlet *et al.*, 2010), which is a relatively low value compared to other plant species. Thus, mechanically speaking, the resistant cross section is therefore close to the apparent section.

In the primary cell wall, microfibrils are extensively reoriented during cell elongation at an early stage of fiber development (Rihouey *et al.*, 2017). In the secondary wall, highly crystalline cellulose fibrils are spirally wound and the microfibrillar angle changes at each of the layers S1, S2 and S3. Nevertheless, within a given layer, the cellulose fibrils have a constant orientation (Rihouey *et al.*, 2017).

The main layer (S2) represents around 80% of the total section, with cellulose fibrils forming an angle of 10° with the axis (Bourmaud *et al.*, 2013). In this layer, the fibrillar orientation corresponds to a S-Twist (Roelofsens, 1951), just as observed in nettle or ramie plant fibers. Inside the S1 layer, the fibrillar orientation corresponds to a Z-Twist (Roelofsens, 1951), but there is a lack of data on the exact MFA for this layer. The innermost S3 or Gn layer is of a transient nature (Rihouey *et al.*, 2017).

To illustrate the differences in fibrillar orientation (S-Twist and Z-Twist), **Fig. 9** compares flax, hemp and nettle fibers.

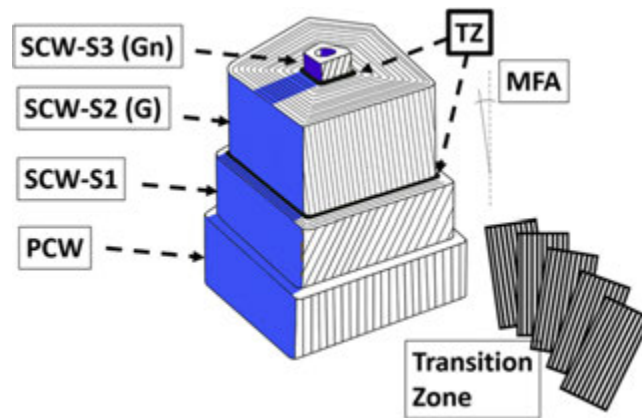


Fig. 8 Schematic drawing of an elementary flax fiber; PCW = primary cell wall; SCW = secondary cell wall with three layers: S1, S2 and S3 (Gn); TZ = Transition zone between S1-S2 and S2-S3. Adapted from Bailey, C., Goudenhooft, C., Gibaud, M., Bourmaud, A., 2018. Flax stems: From a specific architecture to an instructive model for bioinspired composite structures. *Bioinspiration and biomimetics* 13 (2). Available at: <https://doi.org/10.1088/1748-3190/aaa6b7>.

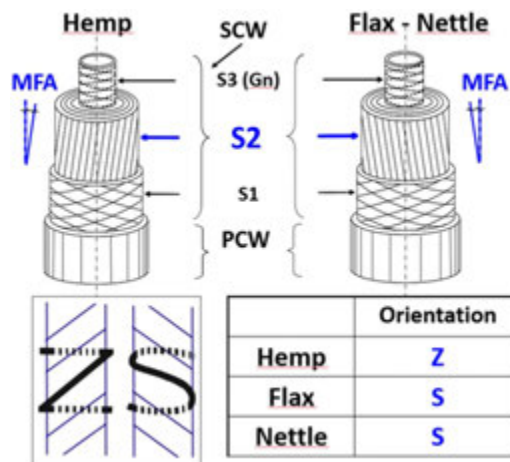


Fig. 9 Fibrillar orientation of the S2 layer. Comparison between flax, hemp and nettle fibers.

Flax Fiber (Cell Wall) Thickening

The cells are initiated by primary growth, from the apical meristem (Ageeva *et al.*, 2005). The fiber initiation is then followed by two main stages: elongation and cell wall thickening. The stage of elongation is particularly outstanding in the case of flax fibers, as their final length can reach up to 65 mm with average values of 25 mm (Gorshkova *et al.*, 2003). These extreme values could be explained by the two-step process of elongation: coordinated growth then intrusive growth (Ageeva *et al.*, 2005). For a deeper understanding of intrusive development, we recommend the following references to the reader (Ageeva *et al.*, 2005; Snegireva *et al.*, 2006). Due to the greater thickness of the flax fiber cell wall compared to other plant species (flax fiber cell walls can reach more than 10 μm , whereas most cell walls are a few microns thick) (Mikshina *et al.*, 2013), the process of fiber thickening has been the subject of many studies. This process results in a uniform cell wall deposition along the fiber length, with several distinct cell wall layers, whose deposition is described hereafter (Snegireva *et al.*, 2010).

At its formation, a flax fiber cell, similarly to other plant cells, is constituted by a primary cell wall (PCW) and fixed with other cells through the surrounding middle lamella (ML). The PCW of flax fibers is a very thin and extensible structure able to withstand the impressive fiber elongation. Subsequently, there is the deposition of the new layers and the first in the S1 of the secondary cell wall, a very thin layer of the SCW (Rihouey *et al.*, 2017). The further deposited layers being responsible for the extreme fiber thickening, and more specifically their denomination, are the subject of debates between authors. It is possible to describe the next additional layer as the major thick layer denominated as the S2 SCW layer or preferentially the G-layer because of its specific properties (Rihouey *et al.*, 2017; Goudenhooft *et al.*, 2019).

Namely, the G-layer is fiber-specific, has a pronounced content of cellulose (up to 90%) with a high crystallinity and a low angle of cellulose microfibrils being almost parallel to the fiber longitudinal axis (with a MFA reaching 8° – 10°). It is also characterized by the absence of both lignin and xylan, a high water content, and its porosity, as well as contractile properties (Mikshina *et al.*, 2013). This remarkable cell wall layer that is the G-layer has led to the term “G-fiber” sometimes used to name the fibers having such a layer (Ibragimova *et al.*, 2017). Lastly, the innermost thin deposited layer is defined as S3 or Gn-layer (Rihouey *et al.*, 2017) (Fig. 8), being in fact a residue of an unmaturing Gn layer whose presence and thickness is variable according to the growing conditions; it is almost absent for fibers that have undergone optimal maturation.

The main stages of fiber thickening are illustrated in Fig. 10 (Arnould *et al.*, 2017; Goudenhooft *et al.*, 2018; Goudenhooft *et al.*, 2019). Table 8 summarizes the characteristics and composition of the different cell wall layers of flax fibers. Observations of the

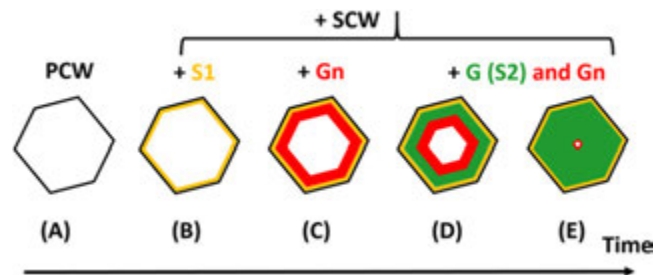


Fig. 10 Diagram illustrating the different stages of the fiber thickening, starting from a cell having only a primary cell wall (PCW) and ending with a fiber having a thick G-layer, a small lumen and a possibly remaining thin Gn-layer. Adapted from Goudenhooft, C., Bourmaud, A., Baley, C., 2019. Flax (*Linum usitatissimum* L.) fibers for composite reinforcement: Exploring the link between plant growth, cell walls development, and fiber properties. *Front. Plant Sci.* 10, (23). Available at: <https://doi.org/10.3389/fpls.2019.00411>.

Table 8 Characteristics and approximate composition of the different cell wall layers of flax fiber

Cell wall layer	Average thickness	Microfibrils orientation	Approximate composition
PCW	0.2 μm	disperse orientation, preferentially 0°	$\sim 25\%$ – 40% Cellulose $\sim 30\%$ Hemicelluloses $\sim 30\%$ Pectins
SCW – S1	0.5 μm	60° – 80°	$\sim 30\%$ – 50% Cellulose $\sim 30\%$ Hemicelluloses $\sim 5\%$ Pectins $\sim 10\%$ – 20% Lignin
SCW – S2 or G	Up to 15 μm or 90% of the total cell wall area at maturity	8° – 10°	$\sim 75\%$ – 90% Cellulose $\sim 15\%$ – 20% Hemicelluloses $\sim 5\%$ – 10% Pectins
SCW – S3 or Gn	0.5–1 μm through thickening	Loosely packed as a heterogeneous structure	Cellulose Hemicelluloses Pectins

Note: Adapted from Goudenhooft, C., Bourmaud, A., Baley, C., 2019. Flax (*Linum usitatissimum* L.) fibers for composite reinforcement: Exploring the link between plant growth, cell walls development, and fiber properties. *Front. Plant Sci.* 10, (23). Available at: <https://doi.org/10.3389/fpls.2019.00411>.

fiber thickening steps through atomic force microscopy (AFM) are shown in the following references (Arnould *et al.*, 2017; Goudenhooff *et al.*, 2018).

Transition Zone

In the literature, the transition zones between the S1 and S2 layers are described, and also between the S2 and S3 layers (Roland *et al.*, 1995; Roland and Mosiniak, 1983; Wang *et al.*, 2013). In these transition zones, microfibrils stay in parallel planes and change their orientation progressively (Fig. 8). Fig. 11 illustrates the transition zones. Fig. 11(A) shows the schematic organization of the plant cell wall in a plant cell wall transition zone (Reis *et al.*, 2006). For more information on this topic, and to see the transition zone organization in 3D (of a spruce tracheid), the reader can consult the following reference (Reza *et al.*, 2014).

This gradual transition (Fig. 11(B)), visible between wall layers (Fig. 11(A)), is a source of inspiration to decrease the risk of delamination in a composite material laminate (Baley *et al.*, 2018; Li *et al.*, 1995). For example without a transition zone, in original industrial plywood the wood layers are oriented in two directions (0° and 90°) (Fig. 11(C)). To avoid the abrupt discontinuity of mechanical properties between a 0° ply and a 90° ply, it is possible, through a bioinspired approach, to change the ply orientation gradually (Fig. 11(B)). An analogy can be established with composite materials; similarly, the abrupt transition between the orientation in a laminated composite represents a main weak point of these materials and a progressive evolution of fiber orientation is a good way to mitigate this drawback.

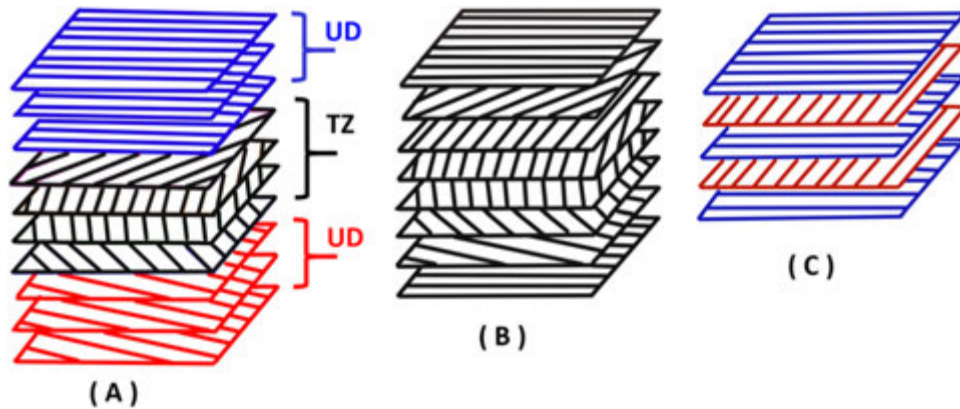


Fig. 11. Stacking principle of unidirectional plies. A: Schematic organization of the cell wall in a transition zone. B: Laminate with a progressive evolution of the angles between each ply. C: Plywood (stacking layers of wood at 0° and 90°).

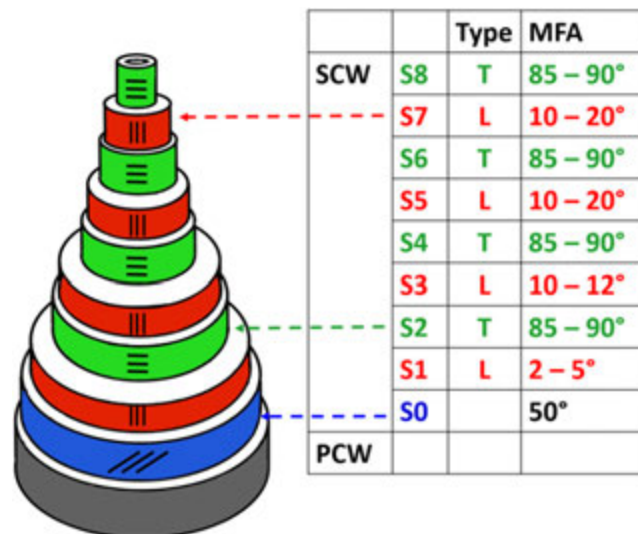


Fig. 12 Model of the polylamellate structure of a thick-walled bamboo fiber. Numbers on the table indicate fibril angles, and letters indicate terminology of wall lamellae. The affixes L and T stand for the almost longitudinal (L) and transverse (T) orientation of the cellulose fibrils in the respective lamellae. Adapted from Parameswaran, N., Liese, W., 1976. On the fine structure of bamboo fibres. Wood Sci. Technol. 10, 231–246.

Bamboo Fiber

In order to illustrate the diversity in the multi-scale organization of plant fibers with a complementary case, this section presents the structure of an elementary bamboo fiber. In bamboo, an elementary fiber has a pentagonal or hexagonal shape and, in the culm, these elementary fibers are assembled into bundles near the vascular cells. Bamboo fiber bundles are distributed densely in the outer region of the culm wall and sparsely in the inner region.

The fiber wall consists of the primary, secondary, and tertiary wall. Specifically, and contrary to previously described flax or wood fibers, the secondary wall is made up of numerous layers of microfibrils with a varied orientation. The inner layer, also called tertiary wall, is often covered by a warty structure (Liese and Köhl, 2015). The cell wall structure of bamboo fibers is quite complex, as shown in Fig. 12, where the outermost layer of the cell wall is the primary wall (PCW), and its microfibril arrangement is reticular and disordered; below the outer layer is a secondary wall (SCW), which is comprised of up to nine (occasionally more) alternately repeated thick, micro-scale layers and thin nano-scale layers (Parameswaran and Liese, 1976). The orientations of thick-layer microfibrils are mostly along the fiber axis (L for longitudinal orientation in Fig. 12)), where the microfibril angle (MFA) shows an increasing trend from the inter-cellular layer to the cell lumen; in the thin layer, the orientations of microfibrils are basically perpendicular to the fiber axis (T for transverse orientation in Fig. 12). The thin layers have a significantly higher lignin content than the thick layers and accordingly exhibit more lignification (Parameswaran and Liese, 1976; Crow and Murphy, 2000). The secondary cell wall transition lamella S0 is not always present. Observations with AFM of the fiber laminate are shown in the following references (Shah *et al.*, 2019).

Conclusion

This article presents the properties, composition and architecture of some plant fibers that can be considered as models. It includes simplifications for pedagogical reasons and does not pretend to be complete, this topic being huge. It is intended for specialists in composite materials wishing to understand the multi-scale structure of this type of natural reinforcement.

Plant fibers are discontinuous and may have one or more functions such as mechanical support and conduction, so there is a diversity of cells. The main criteria for the choice of a reinforcement (with a polymer matrix) are the aspect ratio of the fibers and their mechanical properties. The fiber properties are a function of their composition, architecture and nanostructure. Their intrinsic organization is similar to a laminate of composite materials; at their scale, plant fibers are reinforced by cellulose fibrils which are organized at a nanometric scale, so they can be described as nanostructured composites. Using plant fibers as reinforcement means reinforcing a composite material with fibers that are they themselves composites.

There is a wide range of plant fibers that can be used as reinforcement, wood, flax and bamboo are specifically described here as simplified organizational models. These specific cases enable the understanding of the mechanisms of the reinforcement of plant cell walls through cellulose fibrils arranged in a helix and oriented with a microfibrillar angle.

The term wood fiber describes several types of cells performing various functions in a living tree. In addition, depending on the botanical family of the tree considered, they are not always present. The main type of fiber has been described precisely (architecture, composition, nanostructure) as well as its different cell walls and layers. This multi-scale approach makes it possible to understand the relationships between the stiffness of crystallized cellulose and the wood fiber modulus. Moreover, a plant has the capacity to adapt to its growing conditions. In this context, the architecture of some cells inside a tree trunk evolves, as in the case of tension wood.

In the literature, many scientific articles are devoted to flax fibers, as they have good mechanical properties and a very high aspect ratio. The analysis of their organization enables us to define the notion of a transition zone and to illustrate the thickening of cells over time. It is not just a filling, but a complex construction involving remodeling mechanisms. In addition, the description of an elementary bamboo fiber illustrates another organization of the secondary plant cell wall.

By choice, this article describes the multi-scale structure of a plant fiber that can be used as a reinforcement for composite materials as well as their average properties. Due to lack of space, the relationship between its complex internal organization and its mechanical behavior is not shown in great detail. Furthermore, it would be interesting to consider a plant as a living, composite, hierarchical and optimized natural structure.

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Mechanical Properties of Natural Fiber Composites

Eric Le Bourhis, Institut Pprime, CNRS-Université de Poitiers-ISA-ENSMA UPR, Département Physique et Mécanique des Matériaux, SP2MI, bd M&P Curie, Futuroscope, France

Fabienne Touchard, Institut Pprime, CNRS-ISA-ENSMA-Université de Poitiers UPR, Département Physique et Mécanique des Matériaux, ENSMA 1 av Clément Ader, Futuroscope, Chasseneuil, France

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Nomenclature

d Fiber diameter (m)

F Force (N)

GFRP Glass fiber reinforced plastics

l Embedded fiber length (m)

l_c Critical fragment length (m)

l_f Fragment length (m)

MAPE Maleated polyethylene

MAPP Maleated polypropylene

NFC Natural fiber composite

PA11 Polyamide

PE Polyethylene

PLA polylactide

PP Polypropylene

τ Interfacial shear strength (Pa)

σ_f Fiber strength (Pa)

Glossary

Bast fiber High performance fiber extracted from plant stem.

Glass fiber Synthetic fiber made of E or S glass.

Hydrophilicity/hydrophobicity Attraction (or repulsion) to water.

Interface Interface between the polymer matrix and the fibers.

Interphase transition region between fiber and the matrix.

Natural fiber Fiber of natural origin (animal, plant and mineral).

Tensile strength Resistance to tensile fracture (Pa).

Yarn A thread formed by elementary fibers twisted together.

Introduction and Context

Natural fiber composites (NFCs) allow alternatives to synthetic carbon and glass fiber composites in the context of sustainable materials for a broad range of applications spanning from building structures to aeronautics (Shahzad, 2011; Yashas Gowda *et al.*, 2018; Girijappa *et al.*, 2019; Ramesh, 2019). Natural fibers mentioned in this article are plant-based but it should be noted that animal fibers like cocoon silkworm or spider silk can also be used (Ku *et al.*, 2011; Oushabi, 2019). In fact, natural fibers comprise many different categories, as they may have animal, vegetable and mineral origins. For the sole vegetable origin, subcategories are defined according to their being wood (generally waste materials), extracted from stems, leaves or seeds. The review focuses on high performance natural bast fibers being extracted from plant stem, an example being hemp fiber that is used to illustrate further the topic. So far, the review can be extended to other categories and literature is cited accordingly. The environmental marks of NFCs can be much reduced in terms of energy and emission as compared to conventional ones. In the case of Hemp versus Glass, the marks are lower of an order of magnitude and more (Table 1). As a matter of fact, the natural fiber composites are gaining market share even though the evolution is incremental and glass remains largely dominant (Fig. 1).

NFCs can be classified according to the size of the fibers, namely short (randomly dispersed), medium (using mat or non-woven) and long fiber (unidirectional or woven), together with the associated matrix namely thermoplastic or thermoset. The process used makes the combination of both, bearing in mind that matrices are to be used at temperatures compatible with NFCs. Therefore, polyethylene (PE) or polypropylene (PP) thermoplastic matrix allows for extrusion, compression and injection molding of the composites, while epoxy and polyurethane thermoset composites can be manufactured with resin transfer molding, sheet and bulk molding compound. The overall performance depends on the component properties (matrix and fiber) and on many other interdependent parameters as illustrated in Fig. 2, notably the interfaces as discussed extensively below.

Massive production of polymers in the last century has opened the use of composite materials. Noticeably, natural fibers could have been used originally as shown by the attempts by Ford company already around 1940, who incorporated natural materials to

Table 1 Environmental parameters in the production of 1 kg of fibers

	Hemp fiber	Glass fiber
Power consumption (MJ)	3.4	48.3
CO ₂ emission (kg)	0.64	20.4
SO _x & NO _x emission (g)	2.15	11.7

Note: Shahzad, A., 2011. Hemp fibre and its composites – A review. Journal of Composite Materials 46, 973–986.

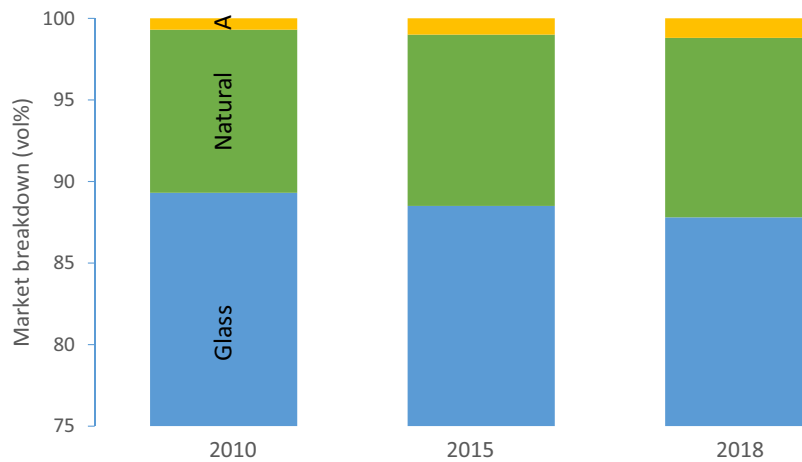


Fig. 1 Market breakdown of fibers over the last decade (A for Aramid). A slight favorable evolution in the direction of natural fiber use is observed. According to Lucintel, E., 2019. Evolution of the composite market by fibre type – 2000–2018. JEC Composites Magazine 1, 39 (special issue #).

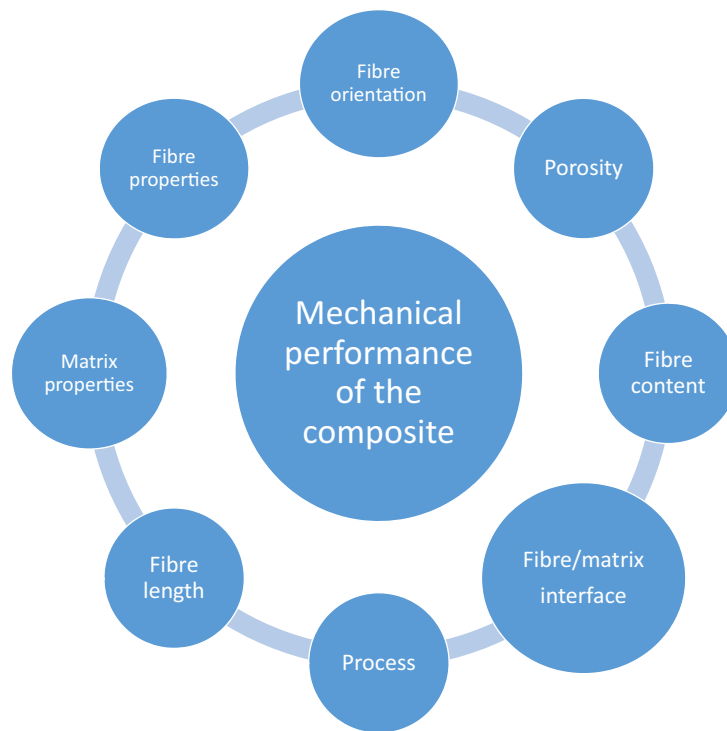


Fig. 2 Influential parameters governing the performance of the composite.

produce organic car body (Evans and Suddell, 2005). Composites have entered all sectors including packaging, building, transportation (automotive, naval, and aeronautics), communication (Evans and Suddell, 2005).

Progressively, NFCs have become used in most of the cited sectors although the mutation remains slow and the market is still dominated by glass fiber composites (Fig. 1). Nonetheless, the massive use of composites causes serious issues in the production (Table 1) and in the disposal of wastes. New regulations are in favor of NFCs either in terms of energy, raw material or disposal. Recycling routes are also to be explored and considered from the design step and here again natural fibers hold great advantage. The natural fibers represent an interesting low-cost and CO₂ neutral resource, as they can be extracted from many different plants like Banana, Flax, Hemp, Jute, some of them being waste products from the agroindustry (Kiruthika, 2017; Evans and Suddell, 2005). Noticeably, the complete sustainability of the composites requires considering bio-based polymer matrices (La Mantia and Morreale, 2011; La Rosa *et al.*, 2014). Thermoplastics exhibit a reduced environmental impact as compared to thermosets, which cannot be recycled. On the other hand, thermosets offer higher mechanical performances.

Table 2 Properties of elementary fibers (order of magnitude)

	<i>Hemp fiber</i>	<i>Glass fiber</i>
<i>Density</i>	1.4	2.5
<i>Stiffness (elastic modulus, GPa)</i>	70	70
<i>Strength (tensile, GPa)</i>	0.7	3
<i>Hydrophilic</i>	yes	no

Table 3 Some applications of NFCs

	<i>Examples of component</i>
<i>Transportation</i>	Aeronautics: Interior panels; Automotive: Insulation, furniture, package shelves, Interior/exterior, door panels, seat backs paneling, bumpers, spoilers; Railway, Interior, door leaves; Naval, hull, panels
<i>Infrastructure, building</i>	Insulation, filling agent, moderate load bearing beam, roof panel, wind mill blade
<i>Packaging</i>	Textile, bag, filter

It is important to emphasize that natural fibers also benefit from a reduction of weight when compared to their glass counterpart (Le Bourhis, 2014) that is of utmost interest for the transportation sector and in particular for the aeronautics (Table 2). Taking again the example of hemp, a factor about two is obtained in terms of density. Still, elementary hemp fiber competes in terms of elastic modulus when tensile strength reduction is the main drawback.

High performance bast fibers extracted from plant stem offer weight saving around 10%–30%. Flax, jute, hemp bast fibers represent the largest sources used by the manufacturers. These plants are cultivated and their production can be renewed on an annual basis to the demand of the manufacturers. Noticeably, the hemp culture is almost pesticide free while this is not the case for the other plants. The performances achieved by the fibers and their NFCs allow producing interior automotive parts as well as exterior components like bumpers, spoilers (Table 3) (Evans and Suddell, 2005; Koronis *et al.*, 2013). As regards the aeronautics, the applications concentrate on the interior (Scarponi *et al.*, 2017). So far, the composites have to meet crucial capability in terms of flammability and impact loading. Structural and infrastructure are also addressed in sectors demanding moderate strength. Load bearing beam, roof panels for tanks, pedestrian bridges have been introduced. Naval applications have been also considered (La Rosa *et al.* 2014; Castegnaro *et al.*, 2017). In particular, La Rosa *et al.* emphasized that the life cycle assessments (LCA) of bio-based polymer composites compare favorably with petroleum-based products (La Rosa *et al.*, 2014).

The two following sections concern mechanical properties both under static and dynamic modes. Mechanical properties addressed here are of utmost importance for the structural or semi-structural applications. A major drawback of natural fiber composites regards the moisture sensitivity. Therefore, this key point is also considered. Last section focuses on fiber/matrix interface that plays a major role in the resistance to fracture and fatigue.

Mechanical Properties

Despite a steadily expanding body of work demonstrating the potential of natural fibers, industry adoption in load-bearing engineering applications is still reluctant due to a general lack of confidence in their mechanical performance, their complex loading behavior and the relative immaturity of research compared to that of synthetic fibers (Mahboob and Bougherara, 2018). In this section, quasistatic and dynamic mechanical properties of NFCs will be discussed.

Quasistatic Behavior of Natural Fiber Composites

In order to evaluate the potential of use of NFCs in engineering applications, many authors have determined the quasistatic behavior of natural fiber composites in tensile, flexural and compressive loadings. For example, Ku *et al.* proposed a review on tensile properties of short plant fibers composites (Ku *et al.*, 2011), Krishnan *et al.* have studied the tensile and flexural behavior of hemp/polyester composites with different kinds of fiber orientations (Krishnan *et al.*, 2020) and Van Vuure *et al.* have compared compressive properties of composites with three different natural fibers: flax, bamboo and coir (Van Vuure *et al.*, 2015). In order to rank the large databank of mechanical properties of NFCs available in literature, Shah proposed different types of selection charts (Shah, 2014). Fig. 3 presents for example a comparison of the absolute and specific tensile properties of various plant fiber composites with glass fiber reinforced plastics (GFRP). It can be seen in Fig. 3 that NFCs, particularly (1) unidirectional thermosets and thermoplastics, and (2) nonwoven thermoplastics, perform exceptionally well against similar GFRPs in terms of both absolute and specific stiffness. However, both the absolute and specific tensile strength of NFCs tend to be lower than that of GFRPs.

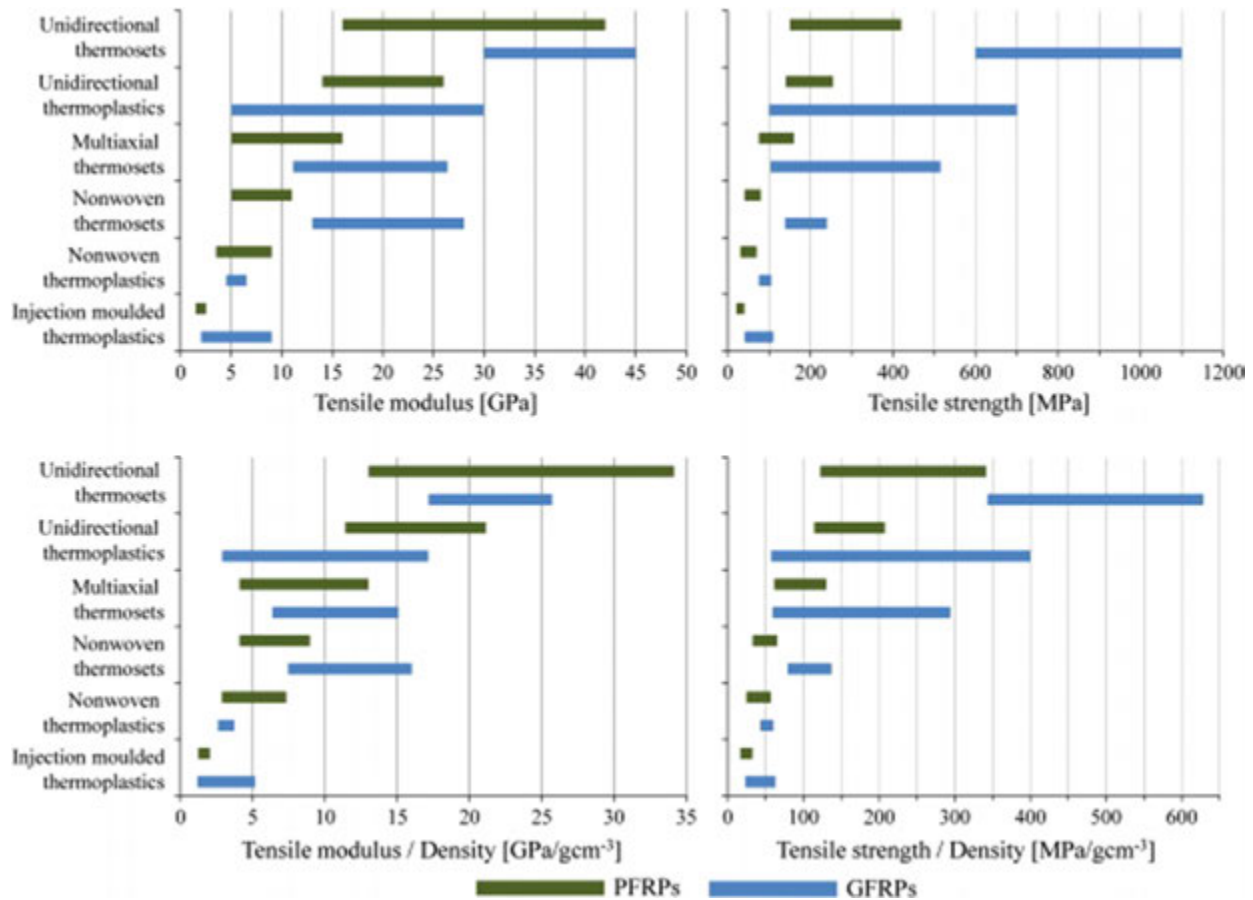


Fig. 3 Comparison of the absolute and specific tensile properties of plant fiber reinforced plastics (PFRPs) with E-glass reinforced plastics (GFRPs). Reprinted from Shah, D.U., 2014. Natural fibre composites: Comprehensive Ashby-type materials selection charts. Materials & Design 62, 21–31, with permission from Elsevier.

Consequently, in terms of tensile properties, it can be said that NFCs may be potential alternatives to GFRPs in stiffness-critical applications, but not in strength-critical applications (Shah, 2014).

Because of the high moisture sensitivity of natural fiber composites, a crucial issue is to assess the influence of water on their mechanical properties. Several studies have shown the loss of rigidity and strength under quasistatic loading after moisture absorption in NFCs (Shahzad, 2011; Azwa *et al.*, 2013; Moudood *et al.*, 2019; Dayo *et al.*, 2020). Some models have been developed to take into account the moisture influence on mechanical behavior of NFCs. For instance, Pan *et al.* have proposed a nonlinear constitutive model for unidirectional natural fiber reinforced composite considering the moisture absorption induced swelling and irreversible energy dissipations (Pan and Zhong, 2014). In order to better understand the degradation phenomena occurring in NFCs submitted to moisture environment, it is necessary to characterise the corresponding damage and deformation mechanisms. Different types of techniques can be used to characterise damage and strain in NFCs: optical and electronic microscopy (Pickering and Aruan Efendy, 2016; Malinowski *et al.*, 2018), acoustic emission (De Rosa *et al.*, 2009; Assarar *et al.*, 2011), digital image correlation (Perrier *et al.*, 2015a; Unlusoy and Melenka, 2019), ultrasonic scanning (Lebaupin *et al.*, 2019; Fischer *et al.*, 2019), infrared thermography (Boccardi *et al.*, 2018; Zhang *et al.*, 2018) and X-ray micro-tomography (Rask *et al.*, 2012; Perrier *et al.*, 2017). Fig. 4 illustrates the ability of X-ray micro-tomography to analyze damage development in NFCs. The damage quantity after tensile failure has been measured in a ± 45 woven hemp/epoxy composite for non-aged and water-aged specimens. Results demonstrated that the relative volume of damage in the water-aged sample is more than twenty times higher than that in the non-aged specimen. This explains the degradation of the mechanical properties for NFCs after moisture exposure.

Dynamic Behavior of Natural Fiber Composites

As regards NFCs dynamic behavior, fatigue and impact loadings are responsible for many, if not most, failures in engineering structures. Indeed, dynamic repetitive loadings (vibration, rotation, wind and wave action, turbulence, pressurization, etc.) at levels much lower than ultimate strengths result in sudden and catastrophic failure due to internal damage accumulation over a period of time. Impact loadings may be induced by falling foreign objects that can cause a drastic reduction in the composite

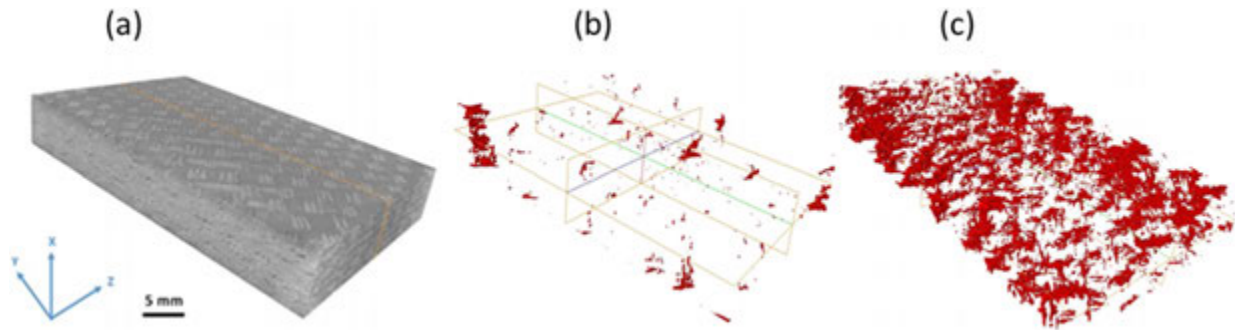


Fig. 4 Damage analysis by X-ray micro-tomography in ± 45 woven hemp/epoxy composite. (a) 3D reconstruction at initial state, (b) and (c) damage after tensile failure in non-aged and water-aged samples respectively.

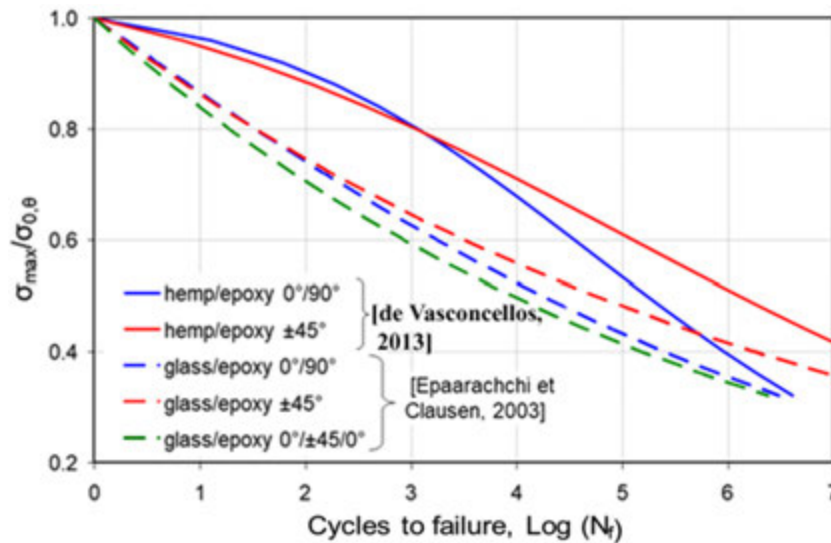


Fig. 5 Comparison of fatigue S-N curves for a hemp/epoxy composite and a glass/epoxy composite.

mechanical properties. Understanding fatigue and impact behaviors of natural fiber composites is thus essential for the efficient and predictable design of load-bearing components.

In their review, Mahboob *et al.* have shown that fatigue testing parameters (e.g., frequency, loading ratio) and material variables (stacking sequence, fiber content) influence longevity (Mahboob and Bougherara, 2018). In the field of short fiber composites, Liber-Knec *et al.* have shown that addition of flax fibers resulted in improvement of fatigue strength of PLA-based composite (Liber-Knec *et al.*, 2015) and Kanny *et al.* have demonstrated that fatigue life of sisal/PP composites was affected due to water content (Kanny and Mohan, 2013). Concerning mat reinforced polymers, it has been shown by Shahzad and Isaac (2014) that hemp/polyester composites exhibit less fatigue sensitivity as compared to glass fiber composites, despite having poorer absolute fatigue strength. The same conclusion can be made for long fiber composites from the work of de Vasconcellos (2013). By considering absolute values, the woven hemp/epoxy composite is not at the level of glass/epoxy composites. But, as it can be seen in Fig. 5, there is a decrease in the rate of fatigue degradation during cyclic loading revealed by the differences in slopes of the S-N curves. The hemp/epoxy composite is less sensitive to fatigue loading than the glass/epoxy composites, which gives an additional advantage to the use of NFCs for industrial applications.

The other type of loading that is crucial for engineering applications is the impact loading. There are many factors controlling impact strength properties of NFCs (Thomason and Rudeiros-Fernández, 2018; Al-Maharma and Sendur, 2019). The impact velocity has a great influence on induced damage, as it has been shown by comparing damages created by a laser shock or by a falling dart impact in a hemp/epoxy composite (Touchard *et al.*, 2017). In laser impacted samples, the induced damage is located near the back face while in mechanically impacted samples, damage appears close to the front face and propagates towards the back face. The natural fiber length plays also a significant role on the impact behavior of NFCs. The comparison of impact damage in short, medium-length and long plant fiber composites has revealed that, despite identical damage mechanisms – i.e., interfacial debonding, matrix cracks and fiber failure –, damage quantity and distribution vary greatly in NFCs made of fibers of different lengths (Malinowski *et al.*, 2018). Another influencing factor on impact properties is the stacking sequence in laminates. Fig. 6

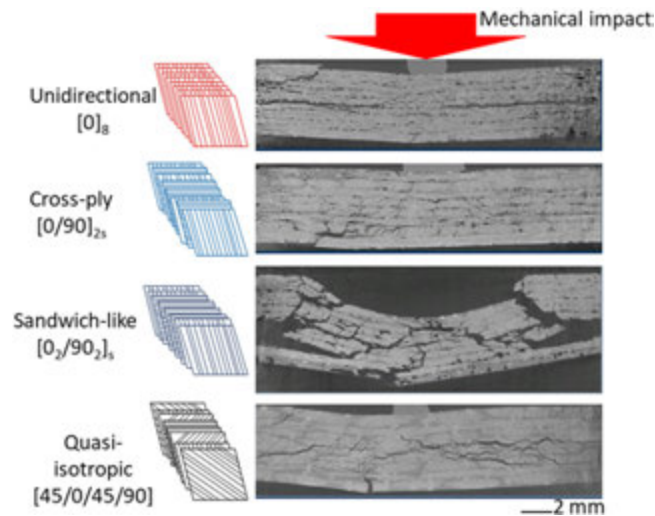


Fig. 6 Micro-tomography images of flax/PA11 composites impacted at 3.6 J for four different lay-ups: unidirectional $[0]_8$, cross-ply $[0/90]_{2s}$, sandwich-like $[0_2/90_2]_s$ and quasi-isotropic $[45/0/45/90]_s$.

clearly illustrates the differences in impact-induced damage for flax/PA11 laminates. This study has been performed in the frame of Lebaupin Ph.D. work (Lebaupin, 2016). Micro-tomography images of flax/PA11 composites impacted at 3.6 J are presented in Fig. 6 for four different lay-ups: unidirectional $[0]_8$, cross-ply $[0/90]_{2s}$, sandwich-like $[0_2/90_2]_s$ and quasi-isotropic $[45/0/45/90]_s$. It is observed that laminates with fibers stacked in the same orientation such as in unidirectional or sandwich-like lay-up are seriously affected by mechanical impact. The induced damage is more significant in these lay-ups than in the two other configurations. This high quantity of damage allows dissipating a large amount of energy during impact loading for these two stacking sequences, but these lay-ups also exhibit poor impact resistance. On the opposite, the micro-tomography analysis shows that damage can be limited by introducing alternated plies or 45° plies in the stacking sequence. Indeed, the quasi-isotropic composite has the smallest induced damage and the highest peak load. It is thus necessary to choose the most suitable lay-up, in terms of impact behavior, for each considered industrial application.

It is also interesting to compare the impact behavior of NFCs with other types of composites. For example, comparisons of hemp/epoxy with glass/epoxy composites (Touchard *et al.*, 2017), and of flax/epoxy with basalt/epoxy ones (Seghini *et al.*, 2020a) have been performed. In both cases, plant fiber composites exhibit a lower peak reaction force value and a better energy absorption capability than glass or basalt fiber composites. The superior capability of NFCs to absorb energy during the impact loading can be related to the high damping properties of plant fibers and to the development of significant damage inside the composites.

All these results show that the mechanical properties of NFCs present interesting aspects compared with classical composites. However, in order to enhance the mechanical strength of NFCs, whether for quasistatic or dynamic loadings, a crucial issue is the need to improve the adhesion quality at the fiber-matrix interface.

Interface

As highlighted above, the properties and performance of a composite rely on its components namely the polymer matrix, the reinforcing fiber and the interface. Latter term has to be defined more precisely since the transition from the fiber to the matrix is not abrupt but encompasses an interphase. The interfacial region controls the stress transfer between the matrix and the reinforcement, which depends on the adhesion level as detailed in the next sections. The improvement of the mechanical properties of the composite may require the pre-treatment of the natural fiber as discussed in Section "Fiber Surface Treatments".

What is an Interface?

The interface appears between the two components of the NFCs namely the fiber and the matrix. The stiff and strong fibers reinforce the polymer matrix on a level depending on the adhesion between both. Adhesion or bonding results from molecular interdiffusion, electrostatic forces, chemical reaction and mechanical interlocking, one of them being dominant (Jacob *et al.*, 2005; Zhou *et al.*, 2016; Latif *et al.*, 2018). As emphasized above, the transition from the fiber to the matrix is not abrupt but consists of a heterogeneous intermediate material. This is particularly evident in Fig. 7(a) where entanglement of fiber and polymer molecules is observed. The transition region or interphase possesses none of the properties of the fiber and the matrix. The nature of the interphase varies with the specific composite system as illustrated in Fig. 7. It is important to note that the interphase is not a distinct phase as it has no clear boundaries. Its extent is difficult to assess although nanoindentation tool has allowed significant

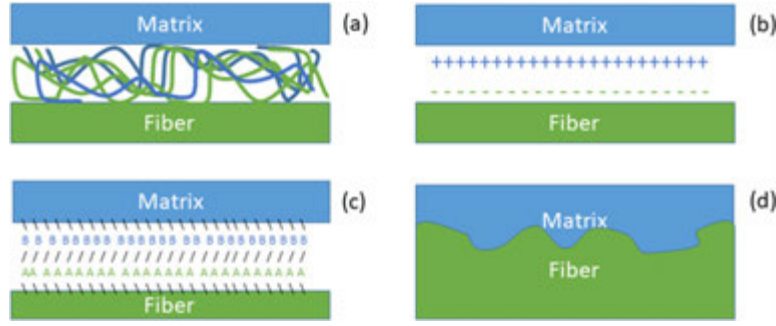


Fig. 7 Fiber-matrix interface with (a) molecular inter diffusion, (b) electrostatic bonding, (c) chemical bonding, and (d) mechanical interlocking.

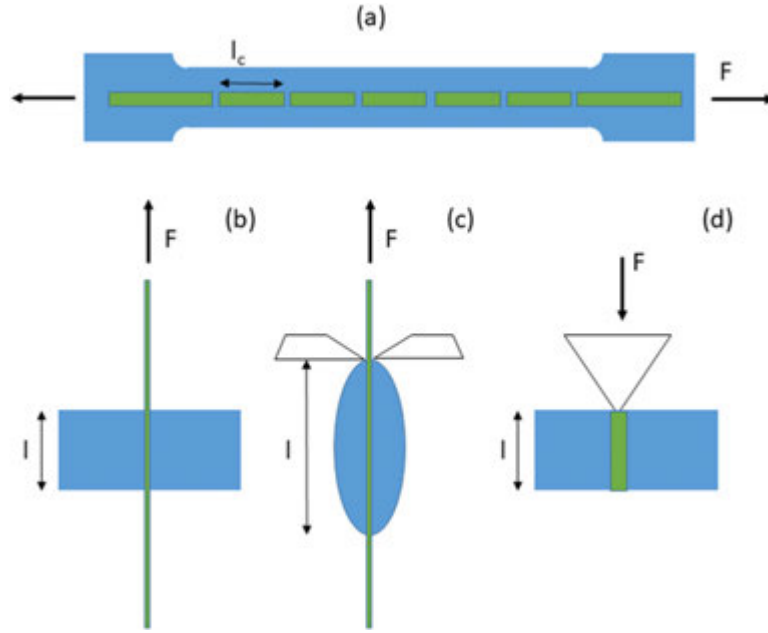


Fig. 8 Micro-mechanical techniques to measure the strength of fiber-matrix interface (a) single fiber fragmentation, (b) single pull-out test, (c) microbond test, and (d) micro-compression.

progress as discussed in Section “Evolution of the Nanoindentation Response at the Interface”. It is believed to be of the order of a few μm . A strong interface allows an efficient stress transfer from the matrix to the fiber and hence higher stiffness and strength values for the composite. However, upon fracture a weaker interface would allow deviation and dissipation. The interfacial strength can be measured by a group of techniques discussed in the next section.

Testing an Interface

The main mechanical characterization techniques used for interface fall in three main categories. In a single fragmentation test, a fiber is embedded in a dogbone matrix specimen and pulled until fragmentation reaches a saturation (Fig. 8(a); Jacob *et al.*, 2005; Zhandarov and Mader, 2005; Zhou *et al.*, 2016). The critical fragment length that can break, l_c , allows determining the interfacial shear strength τ as

$$\tau = \sigma_f d / 2l_c \quad (1)$$

where d is the fiber diameter and σ_f the fiber strength. The second category of test also considers either a single fiber being embedded in a matrix block or in a droplet (Oushabi, 2019). In the first case, the fiber is pulled out, the force F required allowing the determination of the fiber-matrix shear strength τ (Fig. 8(b)):

$$\tau = F / \pi d l \quad (2)$$

where l is the embedded fiber length. In the case of the microbond test with an ellipsoidal droplet (Fig. 8(c)), the force required to debond the fiber against the scissors allows a similar value to be determined with l being the embedded fiber length in the droplet.

It should be noted that one can pass from one equation to the other (Eqs. (1)–(2) and vice versa) when assuming a uniform stress distribution at the interface, a uniform fiber diameter as well as a uniform wetting of the fiber by the matrix (dogbone, block or droplet). So far, tests show large difference in the results of shear strength τ when considering the two main approaches. This is attributed to different stress distribution, due to variable specimen geometry, different behavior at failure and the friction occurring between the fiber and the matrix during a fragmentation and a pull test. In practice, higher values of interfacial strength are obtained with the pullout test than with the fragmentation test (Graupner *et al.*, 2014). In the last category (Fig. 8(d)), we find the indentation approach where a diamond tip is used to push out the fiber. In contrast to the other approach above, actual composite specimens can be used as long as they can be polished carefully without damaging the interfaces. However, indentation push-out has limitation as it is dependent on the assumption made to describe the stress field around the fiber when also the surrounding fibers cannot be neglected. While this method works well with well-defined glass fibers, it becomes tedious with natural fiber bundles. So far, we shall see that using nanoindentation the interphase can be investigated (Section “Evolution of the Nanoindentation Response at the Interface”).

Example With Hemp-Epoxy Interface

The development of woven hemp/epoxy composites for semi-structural applications requires a better understanding of the adhesion mechanisms at the interface between the hemp yarns and the epoxy matrix. In woven composites, yarns are close and interfere with each other. To avoid such influence, single yarn composites can be considered. Different types of analyzes have been performed on such composites and are presented below: measurement at the interface of the evolution of mechanical properties, using nanoindentation and Digital Image Correlation techniques, and determination of the interface adhesion quality, by means of laser shock technique and fragmentation tests.

Evolution of the Nanoindentation Response at the Interface

Interfacial region rules the mechanical transfer between the polymer matrix and the fiber reinforcements. This region or interphase is difficult to scrutinize, its extension depending upon the system. Nanoindentation offers an ideal probe of this interfacial region (Le Bourhis, 2014; Gibson, 2014). Loading-unloading procedure can be adapted to the length scale, the polymer and fiber response while the automatic displacement table offers the possibility of scanning and mapping the region. A single hemp yarn epoxy composite has been elaborated to scan mechanically the interfacial region (Perrier *et al.*, 2016a). Fig. 9 shows a series of Berkovich indents made with a displacement step of 5 μm . The hemp yarn is constituted by several elementary fibers, which can be individual or grouped in bundles (Fig. 9).

Each loading-unloading curve allows to extract locally the elastic response in terms of reduced modulus (Le Bourhis, 2014). A strong contrast of reduced modulus is obtained between the fibers and the matrix with a factor of more than three between both. The data are rationalized once plotted as a function of the distance from fiber edges (Fig. 10). Such a representation allows detecting the intermediate region between the fiber and the epoxy matrix. In the present case, a progressive change of the elastic response is detected with an interphase region being about 2 μm in size. The interphase might be difficult to detect as illustrated in the case of cellulose fiber reinforced polypropylene (Lee *et al.*, 2007). These authors emphasize that the convolution of the probe with the interface is to be considered carefully. It is interesting to note that Nair *et al.* also worked on cellulose fiber reinforced PP composites (Nair *et al.*, 2013). The interphase region in the specimen was observed to be dependent on the matrix used. A 0% maleated polypropylene (MAPP) showed a narrow interphase with a steep modulus gradient in, while the use of MAPP significantly increased the interphase thickness, resulting in a more gradual change in modulus from fiber to matrix. As discussed in more detail in Section “Fiber Surface Treatments”, maleated matrices offer indeed improvement of mechanical properties of related composites.

Strain fields measured by digital image correlation (DIC)

The principle of DIC technique is based on a unique random pattern, recorded twice, once before loading the sample and once after the sample is deformed. The first picture taken at the initial state is the reference picture, which is compared to a second one

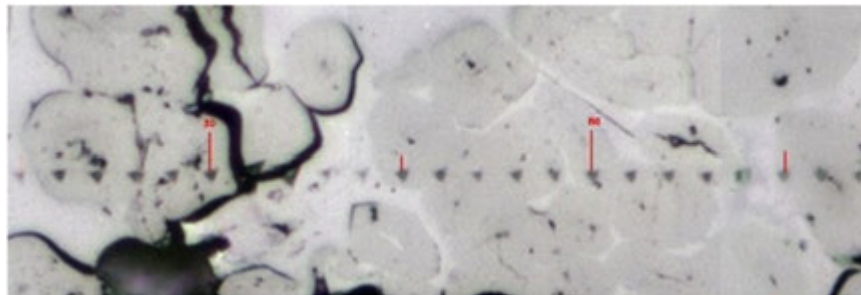


Fig. 9 Nanoindentation scan made in a single hemp yarn epoxy composite as observed by optical microscopy. The distance between indents is 5 μm .

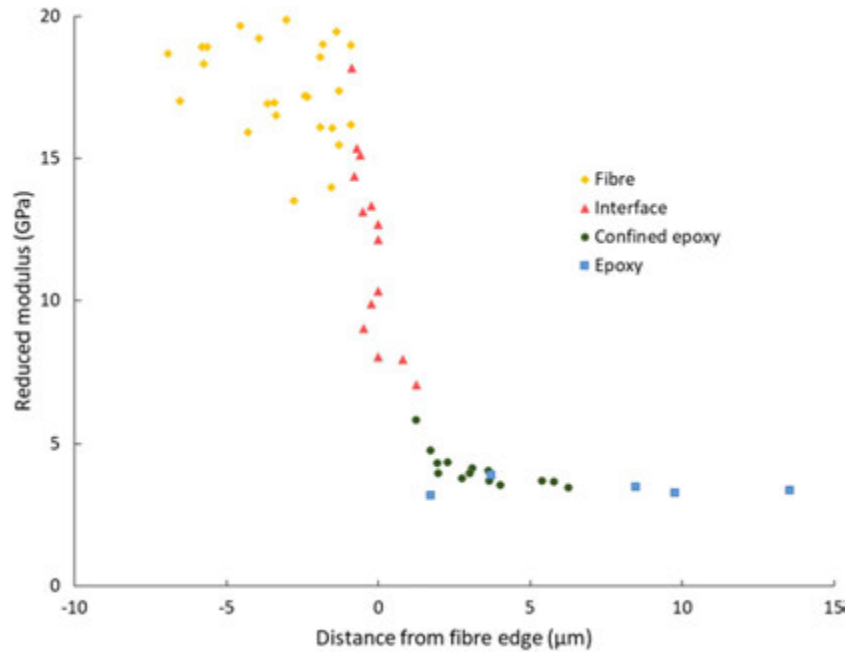


Fig. 10 Reduced modulus for hemp/Epoxy composite versus distance from fiber edge (set to 0) as extracted from the nanoindentation loading-unloading curves. The transition is not abrupt but reveals an interphase.

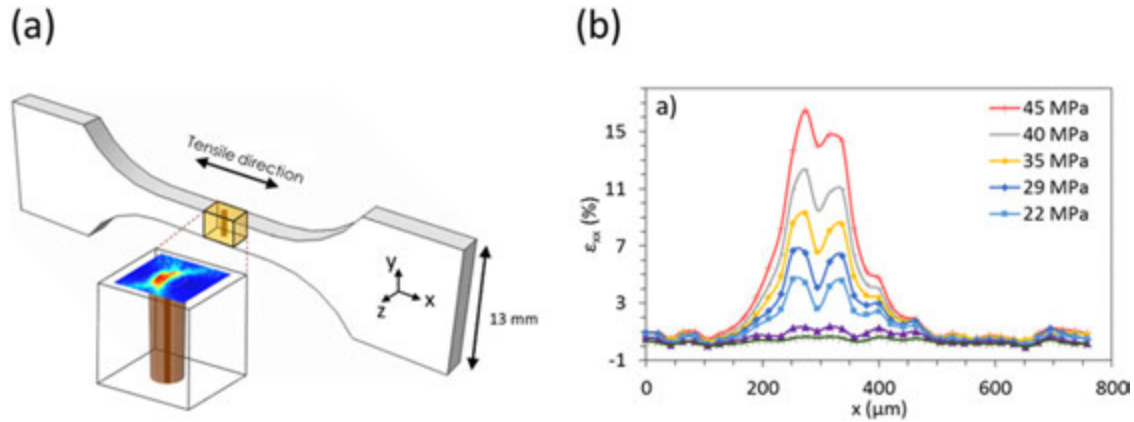


Fig. 11 (a) Single yarn composite samples with hemp yarn oriented at 90° in regard to the tensile direction. (b) Longitudinal strain values along line A measured by DIC at different applied stress levels on the edge of 90° oriented yarn specimens in a single hemp yarn/Epilam sample.

taken at a deformed state. The first picture is divided into small sub-windows, each one being a measurement point characterized by its gray level distribution. The analysis consists in seeking, for each sub-window of the reference image, the most similar one in the deformed image (in terms of spatial distribution of gray levels), using a correlation function. In this study, single hemp yarn specimens with hemp yarn oriented at 90° in regard to the tensile direction were prepared by laying a mixture of paint and particles of 200 nm diameter (Fig. 11(a)). This made it possible to obtain a fine random pattern on the material surface. The thickness of the paint layer was about 33 μm . In order to perform measurements with high spatial resolution, tests were carried out with a micro tensile tester placed under an optical microscope (Perrier *et al.*, 2016b).

Fig. 12 shows longitudinal strain maps measured on the edge of a single yarn composite, with the yarn tilted of 90° . For the different applied stress levels, the same color code has been used; in this way, the evolution of strain is easily showed. The measurements show that strain distributions are highly heterogeneous, with their values increasing with the applied stress level. Strains are localized inside the section of the yarn (represented by the dotted circle). For an applied stress of 45 MPa, which corresponds with 2.31% of macroscopic strain, the maximum strain value measured by DIC is nearly 17%.

The evolution of the measured strain can be more accurately described by plotting in a same graph the strain values for different stress levels of the specimen during a tensile test. Fig. 11(b) shows the longitudinal strain values along the line A for different applied stress levels. It can be seen that the strain increases progressively and is localized at the yarn section. These results allow to

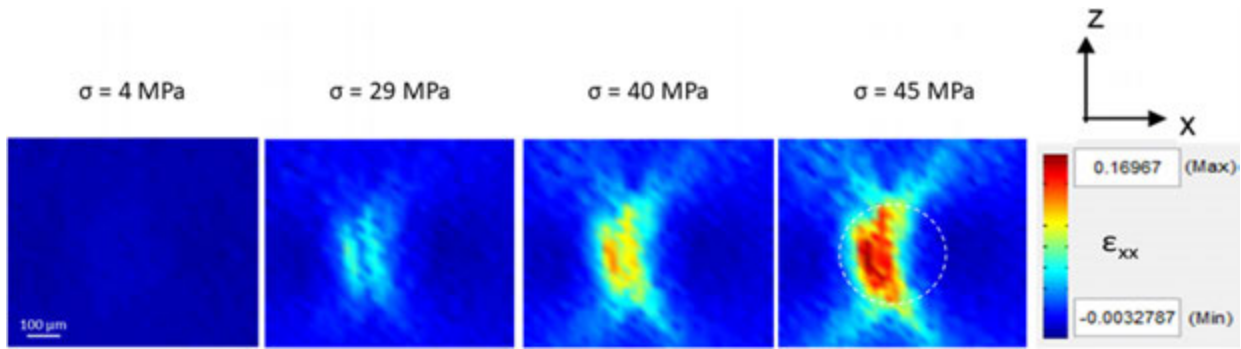


Fig. 12 Evolution of strain fields ϵ_{xx} obtained by DIC versus the applied stress on the edge of a 90° oriented single hemp yarn/Epilam sample.

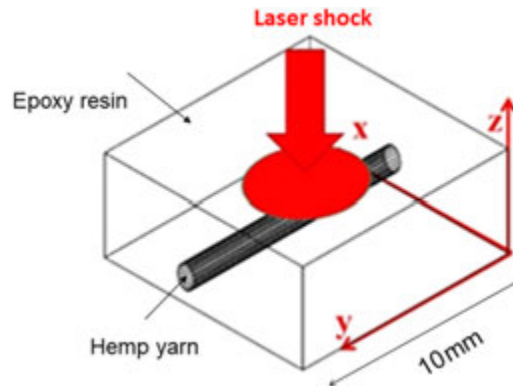


Fig. 13 Composite samples for laser shock tests: single hemp yarn embedded in epoxy resin.

investigate the local strain mechanisms at the yarn/matrix interface, showing the strong heterogeneities in strain fields which develop at the interface.

Interface bonding quality tested by laser shock

The laser shock wave method can create a short but intense internal loading into the shocked sample. Thanks to the wave transmission/reflection through the irradiated material, localized tension in the loading direction can be generated. The laser shock adhesion test (LASAT) technique can be used for example to test the adhesion quality of bonded composite materials (Ecault *et al.*, 2016).

This method has also been used in order to study the adhesion between a single hemp yarn and an epoxy matrix (Perrier *et al.*, 2015b). Samples were made of a single hemp yarn embedded in epoxy resin. Each specimen was cut into square samples, with dimensions of about $10 \times 10 \text{ mm}^2$ and thickness about 3 mm. In these samples, the single yarn was centered in the x direction and positioned along the z direction closer to the back face than to the front face (Fig. 13). Front faces were covered with aluminum paint.

After the shocks, all samples were recovered and damage micromechanisms have been analyzed by microscopic observations. With transmitted light, it was possible to observe single-yarn samples through the matrix (Fig. 14). Two different types of damage induced by laser shock have been observed: resin cracks appear only for high laser intensity levels, and specific cone-shaped interfacial damage appears for lower intensity values. At the yarn-matrix interface, the observed micro-damage is quite unusual: it initiates at one interfacial point, probably a small defect at the interface, and propagates from the yarn surface to the resin. The central axis of the resulting three-dimensional cone shaped cracks is perpendicular to the yarn. It has been observed that this interface damage appears above a given laser intensity value, which corresponds to the damage threshold of the interface. Its quantity and size increase with the laser intensity levels.

The second type of damage consists of large cracks in the resin, localized between the yarn and the specimen back face, as shown in Fig. 14. These cracks are due to the boundary conditions induced by the sample holder, leading to a bending phenomenon in the specimen for high laser intensity level. As for interface damage, these resin cracks appear above a given level of the laser intensity, corresponding to resin damage threshold. These damage thresholds, given in terms of laser intensity levels, are directly linked to the corresponding stress values induced in the material by the shock wave propagation. Therefore, it allows characterizing the mechanical adhesion quality at yarn/matrix interface and the resin damage threshold. It has been determined that hemp/epoxy interfacial damage starts at 0.31 GW/cm^2 and resin cracks start at 0.57 GW/cm^2 (Perrier *et al.*, 2015b). A numerical simulation by finite elements has also been performed to get further insight into laser shock wave propagation in such samples.

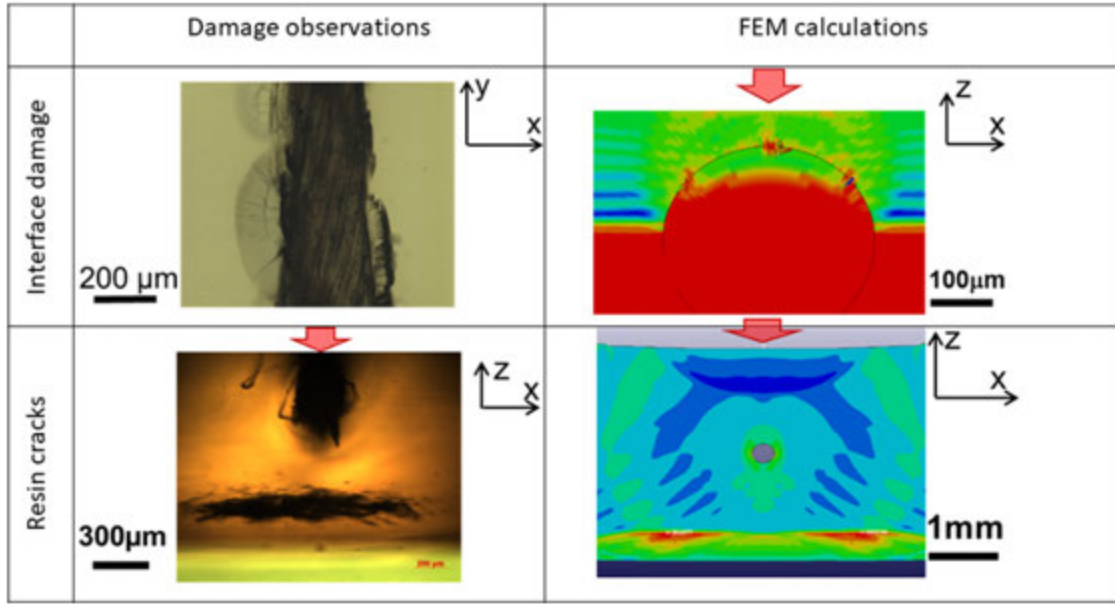


Fig. 14 Damage observations in single hemp yarn/epoxy composite samples after laser shock tests and corresponding σ_{zz} fields obtained by FEM calculations.

The qualitative finite element model (FEM) represents a slice of a sample in the (x-z) plane, with a thickness of one element of 1 μm . The diameter of the hemp yarn section is 300 μm . The laser shock has been simulated by a loading with a Gaussian-like shape in time and a maximum pressure level of 1 GPa. As observed in Fig. 14, FEM calculations lead to three damaged zones at the yarn/matrix interface, which can be related to the micro-damage experimentally observed. Moreover, calculations also show an area of over stresses σ_{zz} near the sample back face, corresponding to the observed resin cracks. These preliminary results demonstrate the ability of the laser shock test to study and quantify the mechanical quality of yarn/matrix interface, which is needed to help design of such composites.

Fragmentation tests

Fragmentation tests are usually performed on elementary glass or carbon fibers embedded in polymer matrix (Zheng *et al.*, 2020; Yao *et al.*, 2017; Swaminathan *et al.*, 2018). Nonetheless, this test method applied to natural fiber composites suffers from several limitations. The length of elementary hemp fibers, about 15 mm, is very small for elaborating a dogbone composite specimen. Moreover, in structures made of woven hemp composites, the load-bearing interface is the yarn-matrix interface. Therefore, the interfacial adhesion should be assessed at the yarn scale. A special mold was developed to elaborate composite samples with a single hemp yarn aligned in the loading direction. Single yarn composite specimens were manufactured with resin injection molding technique using EPOLAM 2020 epoxy resin from Axson. The specimens were dogbone shaped, with a total length of 50 mm. When applying a tensile loading to these samples, the load is transferred through the matrix into the yarn by means of shear stress at the interface. Yarn failure occurs when this transferred stress reached the tensile strength of the yarn. The yarn will continue to fracture into shorter length fragments as the load increases, until the yarn fragments are so small that the tensile stresses induced in the yarn can no longer reach the yarn tensile strength. At this point, a state of saturation is reached, and the fragmentation process ceases. It is thus possible to measure the fragment lengths, l_f , by optical microscopy and to determine the corresponding critical fragment lengths (Eq. 3) (Ohsawa *et al.*, 1978):

$$l_c = \frac{4}{3} l_f \quad (3)$$

An example of hemp-epoxy sample after fragmentation test is presented in Fig. 15. At the end of the test, after the sample failure, several fragments are visible in the gauge length.

The mean value of l_c determined for hemp/epoxy interface is 3.0 ± 0.15 mm, which gives a mean interfacial shear strength (τ) value (Eq. 1) of 14.4 ± 4 MPa (Barbière *et al.*, 2018). Moreover, X-ray micro-CT observations with a very fine resolution (1.5 μm per voxel) allowed the observation of interfacial debonding in the vicinity of the yarn fragment extremity (Fig. 16).

Two different zones are revealed in Fig. 16. The healthy zone, far from the yarn fragment extremity, where the cross-section shows that the interface is undamaged (Fig. 16(b)), and the debonding zone, near the yarn fragment extremity, where damage is present all around the yarn (Fig. 16(c)). These observations confirm that the load-bearing interface is the yarn/matrix interface.

Fiber Surface Treatments

Natural fibers are mostly formed by cellulose, hemicellulose and lignin (Kabir *et al.*, 2012; Liu *et al.*, 2017). Cellulose fibers are highly polar materials and their compatibility with very apolar matrices is highly problematic. Furthermore, due to their polar nature they

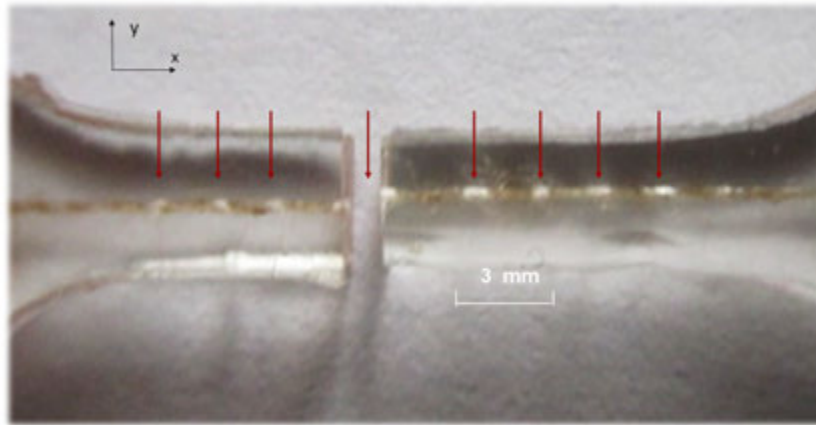


Fig. 15 Single hemp yarn-epoxy composite sample after fragmentation test.

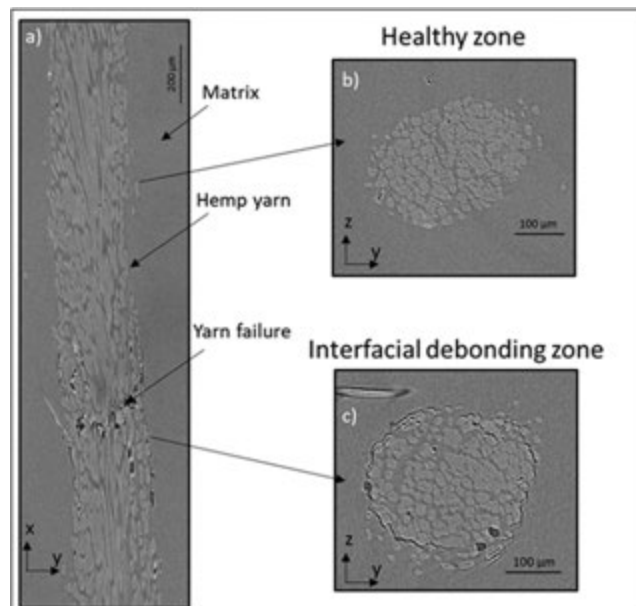


Fig. 16 Micro-CT observations of a single hemp yarn-epoxy composite sample after fragmentation test in the vicinity of a yarn fragment extremity: (a) longitudinal view of a yarn failure zone, (b) cross section in the healthy zone, (c) cross section in the debonding zone.

are very hydrophilic materials, which also poses restrictions on their use in some applications (Zafeiropoulos *et al.*, 2007). A possible solution is to improve fiber-matrix interface by using compatibilizers and adhesion promoters (Shahzad, 2012). Their hydrophilic character is a major drawback as it represents the risk of lesser interfacial contact with the polymer matrix, water uptake on long duration use and microorganism growth (Liu *et al.*, 2017). Those deleterious processes affect the long-term applications of the NFCs and have to be prevented. The hydrophilic character has its origin in the presence of hydroxyl ($-OH$) groups on the cellulose component of the fiber and carboxylic acid ($-COOH$) on the hemicellulose one. Treatments using mercerization, silanization, acetylation, allow reacting with or replacing the ($-OH$) groups producing fibers more resistant and more compatible to the polymer matrix (Kabir *et al.*, 2012; Mohit and Arul Mozhi Selvan, 2018; Oushabi, 2019). Moreover, the hydrophilic character being modified, natural fibers are better wet by the hydrophobic polymer matrix. Oushabi reviewed the literature and showed that the pull-out interfacial shear strength was largely improved upon such treatment regardless of the fiber-matrix system (Oushabi, 2019). Interestingly, this improvement is shown to be correlated with SEM that revealed matrix residues on the fiber after the test (Oushabi, 2019). On the other hand, maleated matrices like polyethylene or polypropylene (MAPE & MAPP) offer a better coupling to the natural fibers (Latif *et al.*, 2018; Oushabi, 2019) correlated with the presence of a thicker interphase region (Nair *et al.*, 2013). In the work of Seghini *et al.*, it has also been demonstrated that oxygen and tetravinylsilane plasma treatments (pp-TVS) allow enhancing the adhesion of flax yarns with epoxy matrix (Seghini *et al.*, 2018, 2020b). By using fragmentation tests on single flax yarn composite samples, it has been shown that flax yarns with pp-TVS deposition after a 100 W plasma oxygen pretreatment lead to a τ value twice

as large as the one obtained with untreated flax yarns. Overall, these approaches promote interfacial adhesion and load transfer. High quality composites can then be obtained (La Mantia and Morreale, 2011).

Conclusion and Prospects

Composite materials have been considerably developed in search of weight gain. Conventional composites become challenged by always improving natural composites. Also, immense applications of NFCs are being opened in the framework of sustainability. NFCs represent strong alternatives of conventional composites used in different important industries. Applications cover transportation, infrastructures and packaging. Some of them are very demanding and it is important to assess the mechanical performance notably the toughness and fracture behavior of these composites. A particular attention has been paid to quasistatic and dynamic (notably fatigue) behaviors where NFCs present interesting performances compared with conventional composites. The main drawback of the use of natural fiber remains its hydrophilicity, which is being circumvented thanks to preliminary treatments. Such treatments allow improving fiber durability against moisture as well as wettability of the polymer matrix. The obtained NFCs show stronger interface and mechanical performance. Albeit these promising aspects, green composites based on natural fiber still need to undergo fundamental investigations to assess their capabilities and potential as conventional composites replacement. The research field is very active as illustrated in the review and will further open new opportunities for NFCs applications.

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Multiscale Tribo-Mechanical Behavior of Natural Fiber Composites

Faissal Chegdani, Arts et Metiers Institute of Technology, Mechanics Surfaces and Materials Processing, HESAM University, Châlons-en-Champagne, France

Mohamed El Mansori, Arts et Metiers Institute of Technology, Mechanics Surfaces and Materials Processing, HESAM Université, Châlons-en-Champagne, France and Texas A&M Engineering Experiment Station, Institute for Manufacturing Systems, College Station, TX, United States

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Introduction

Natural fiber composites (NFC) are arousing the interest of automotive and aerospace industry thanks to many economic, ecological, and technical benefits that natural fibers provide to the composite applications (Dittenber and GangaRao, 2012; Faruk *et al.*, 2012; Mastura *et al.*, 2018; Ramesh *et al.*, 2017; Yan *et al.*, 2014). Therefore, many research works have addressed the manufacturing processes of these ecofriendly materials to investigate the possibility to substitute glass fiber composites in the composite industry (Bos *et al.*, 2006; Goutianos *et al.*, 2006; Jiang *et al.*, 2017; Shah, 2013; Shalwan and Yousif, 2013).

Machining processes, which are unavoidable operations for industrial parts made with long fiber composites, were also investigated to understand the cutting behavior of natural fibers and optimize the machined surface quality of NFC materials (Chegdani *et al.*, 2018a, 2016; Chegdani *et al.*, 2015a,b; Chegdani and El Mansori, 2019, 2018; Chegdani and Mansori, 2018; Lotfi *et al.*, 2019; Nassar *et al.*, 2017; Rajmohan *et al.*, 2019; Roy Choudhury *et al.*, 2018; Vinayagamoorthy and Rajmohan, 2018). It has been shown that the machinability of NFC is highly sensitive to the small variations of material and process parameters. The specific cutting behavior of natural fibers is caused by their multiscale complex structure that differs from nanoscale to macroscale (Baley, 2002; Charlet *et al.*, 2007; Marrot *et al.*, 2013). In order to control the machinability of NFC materials, the tribomechanical behavior of natural fibers within the composite should be investigated and understood at each scale level of the natural fibrous structure.

This article proposes a multiscale tribomechanical study on flax fiber reinforced polypropylene composites. The aim is to explore both the mechanical and the tribological responses of flax fibers and polypropylene separately and at different contact scale levels. This will give to the reader an advanced knowledge about the scale effect on the tribomechanical behavior of natural fibers within the composite materials.

Multiscale Structure of Natural Fibrous Reinforcement in Composites

Fig. 1 illustrates the multiscale structure of NFC materials from macroscale to nanoscale. The macroscopic scale covers the overall composite structure that contains the natural fibrous reinforcement and the polymer matrix (Fig. 1(a)). The mesoscopic scale distinguishes the technical fiber that includes some elementary fibers gathered together naturally with pectic interfaces (Fig. 1(b)). At microscale, each elementary fiber is structured with a stacking of cellulosic cell walls (Fig. 1(c)). The important cell wall is S2 that covers the main fiber volume and controls the fiber properties (Baley, 2002). As shown in Fig. 1(d), the cell wall S2 is itself a composite material at nanoscale with cellulose microfibrils embedded in natural amorphous polymers of hemicellulose, lignin, and pectin (Baley, 2002; Charlet *et al.*, 2007). Cellulose microfibrils are oriented toward the fiber axis with an angle " θ " called microfibrillar angle.

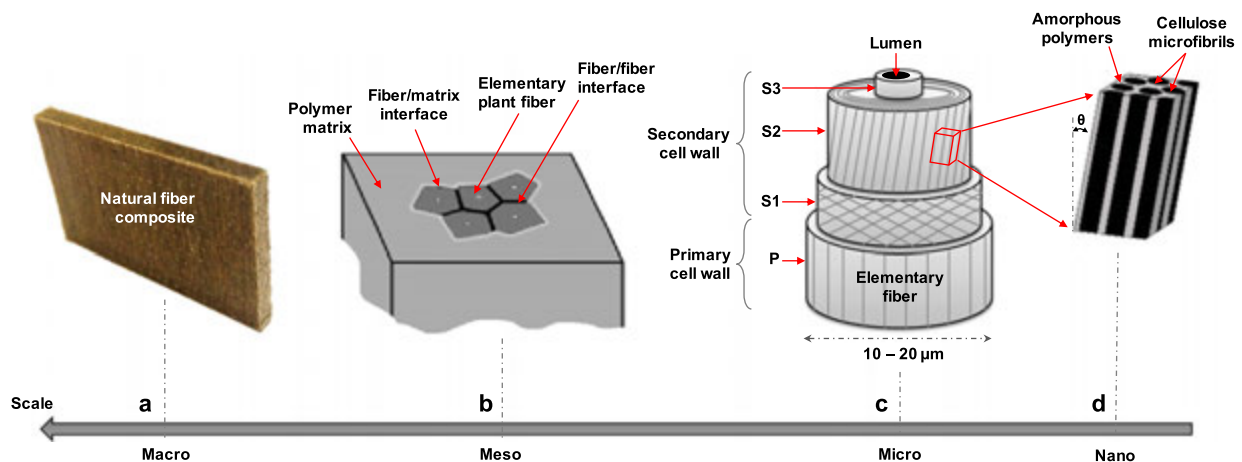


Fig. 1 Multiscale structure of natural fiber composites from macroscale to nanoscale.

Fig. 1 demonstrates that NFC materials have different compositions that vary by changing the scale level. This finding is should be considered to investigate the behavior of NFC during high tribomechanical solicitations such as machining. The next sections will address the contact scale effect on the tribomechanical response of NFC.

Experimental Procedure

NFC Samples

NFC samples used in this study are composed of unidirectional flax fibers as reinforcement and polypropylene (PP) as polymer matrix as shown in **Fig. 2(a)**. The worksurface is perpendicular to the fiber orientation in order to perform the tribomechanical experiments on the cross-sections of flax fibers as shown in **Fig. 2(b)**. It can be seen that flax fibers present a high variability in terms of shape and diameter which a high variability on their mechanical properties obtained from literature by mechanical tensile tests (Dittenber and GangaRao, 2012).

Tribomechanical Tests Approach

The tribomechanical method used in this study is based on nanoindentation and scratch-test experiments. The choice of these two tribomechanical techniques is justified by the fact that both mechanical and tribological experiments should be performed on flax fibers and PP matrix separately in order to reveal the tribomechanical response of each composite phase.

Nanomechanical characterization is used to measure and evaluate numerous mechanical properties of materials, including modulus, hardness, fracture toughness, wear resistance and friction coefficient. Nanomechanical characterization, as well as visualization of surface topography, provides crucial information concerning the performance of materials. Nanoindentation is commonly used to characterize the local stiffness of materials by determining the elastic modulus and hardness. On the other hand, acquiring quantitative force and displacement data in the lateral direction leads to the quantification of friction coefficient, scratch resistance and wear parameters.

Two different techniques for nanoindentation and scratch-test have emerged. These are Atomic Force Microscopy (AFM) and Rigid Probe Microscopy (RPM) as shown in **Fig. 3**. Nanoindentation and nanoscratching with AFM technique are based on the cantilever deflection when the tip indenter is in contact with the material (**Fig. 3(a)**). The cantilever deflection is measured using a laser sensor. This characterization method requires small contact areas in order to avoid the inaccuracy of measurements caused by the own deflection of the cantilever. Therefore, smaller tips indenter radii ($r < 100$ nm) can be used to indent the sample surface, and it is possible to produce low force indentations over the desired region with well-controlled position accuracy (Monclus *et al.*, 2010). For the RPM technique, the tip indenter is directly related to a rigid probe that is normal to the surface as shown in **Fig. 3(b)**. This configuration avoids the cantilevered motion and allows the use of higher loadings and higher tip indenter radii comparing to the AFM technique.

The AFM method has been used in this study with "Dimension Edge™" instrument from "Bruker®". Berkovich diamond tip indenter is considered with a small tip radius ($r = 40$ nm). The tip indenter is related to a steel cantilever that has a spring constant of 450 N/m.

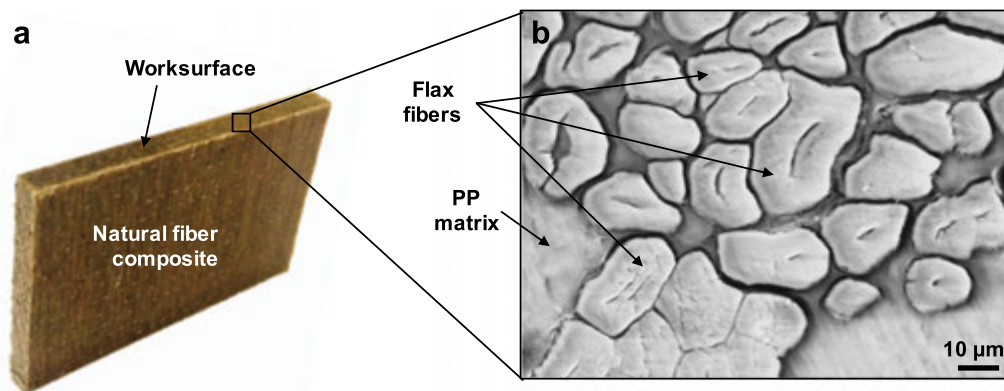


Fig. 2 (a) Photographic image of unidirectional flax fiber composite sample. (b) SEM image of the worksurface of unidirectional flax fiber composite sample.

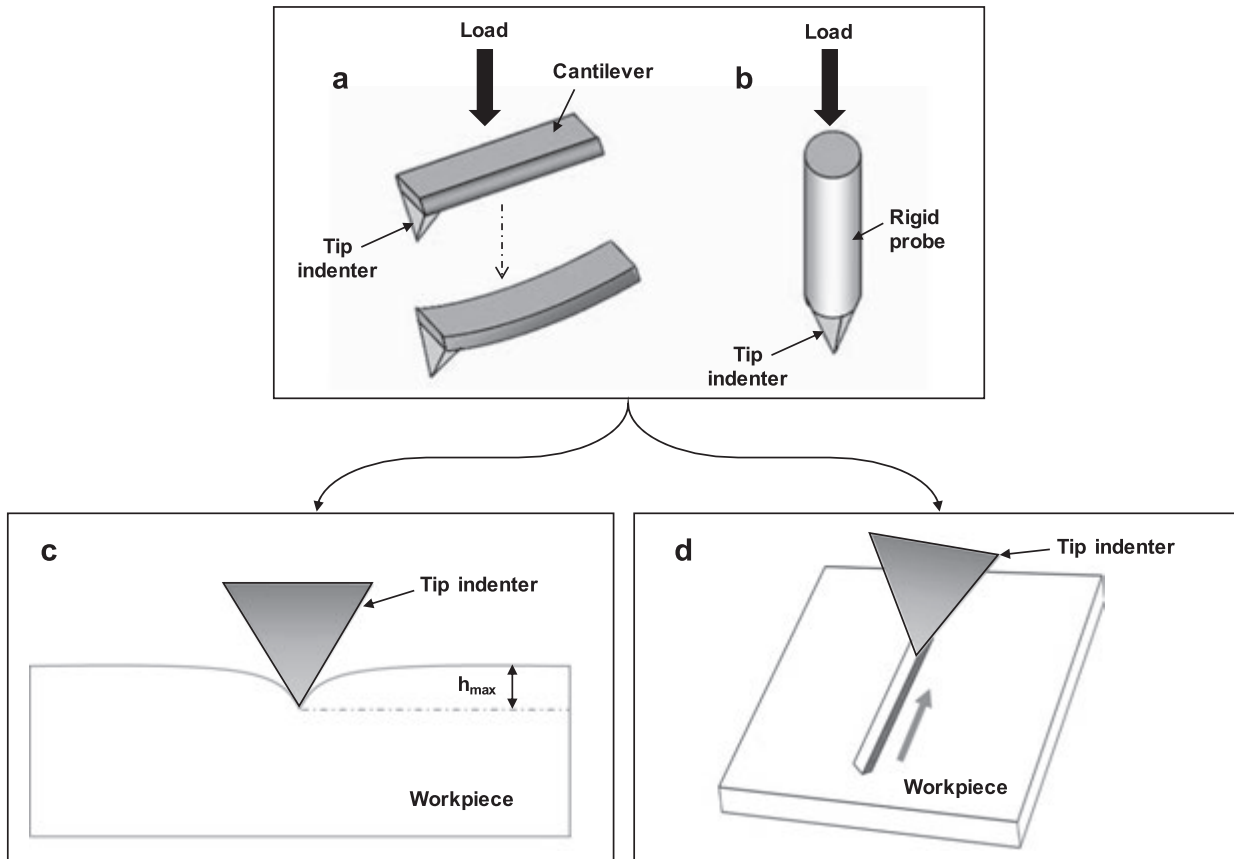


Fig. 3 Schematic illustration of (a) AFM indenter, (b) RPM indenter, (c) Nanoindentation technique, and (d) Scratch-test technique.

The RPM technique has been performed with two instruments:

- The commercial tribomechanical instrument “Nanoindenter XP” from “MTS Nano Instruments®” equipped with a Berkovich diamond tip indenter that has a tip radius of ~ 400 nm.
- The commercial tribomechanical instrument “TI-950” from “Hysitron®”. Two tip indenters have been tested: Berkovich diamond tip indenter and Berkovich Sapphire tip indenter that have a tip radius of 100 nm and 150 nm, respectively.

The aim of this instrument’s choice is to investigate a large range of tip indenter radii and evaluate the effect of the geometrical contact scale.

Nanoindentation analysis

Nanoindentation method involves the normal contact of an indenter on the worksurface and its penetration in this surface to a specified load or depth as illustrated in **Fig. 4(a)** (Bourmaud and Pimbert, 2008). The load is measured as a function of penetration depth as shown in **Fig. 4(b)**. From this load–penetration curve, the pertinent parameters for the analysis are the maximum displacement (h_{max}), the maximum load on the sample (F_{max}), and the contact stiffness (S) which is the slope of the tangent line to the unloading curve at the maximum loading point (see **Fig. 4(b)**).

In the case of Berkovich tip indenter, the model of Oliver & Pharr (Oliver and Pharr, 1992) is generally used to calculate the elastic modulus of each material using the parameters extracted from load–penetration curve of nanoindentation. This model assumes the elastic behavior as the basis foundation of this calculation procedure (Doerner and Nix, 1986; Oliver and Pharr, 1992). The Oliver & Pharr method consists on computing the contact depth (h_c) which is dependent on the material deformation and the tip shape as shown in **Fig. 4(a)**. h_c can be calculated using the Eq. (1). ε is a constant related to the tip geometry (0.72 for Berkovich tip (Bourmaud and Pimbert, 2008)). The projected contact area (A) can be calculated using the Eq. (2). Then, the reduced elastic modulus is obtained using the Eq. (3) where β is a constant related to the tip geometry (1.034 for Berkovich tip (Bourmaud and Pimbert, 2008)). Finally, the elastic modulus of the indented material can be calculated with the Eq. (4) where E_i and ν_i are respectively the elastic modulus and the Poisson coefficient of the tip indenter. ν is the Poisson coefficient of the indented material.

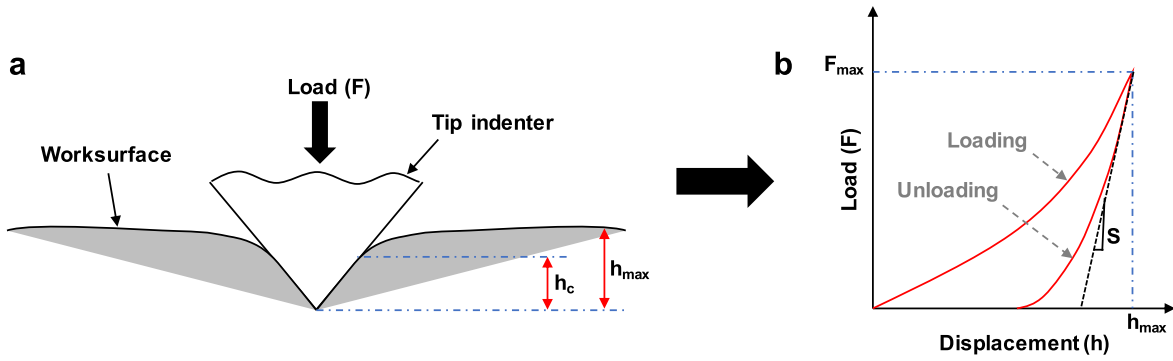


Fig. 4 (a) Schematic illustration of the contact between the tip indenter and the worksurface. (b) Typical load/displacement curve generated by nanoindentation tests.

$$h_c = h_{max} - \varepsilon \frac{F_{max}}{S} \quad (1)$$

$$A = 24.56 \times h_c^2 \quad (2)$$

$$E_r = \frac{S\sqrt{\pi}}{2\beta\sqrt{A}} \quad (3)$$

$$\frac{1}{E_r} = \frac{(1 - \nu^2)}{E} + \frac{(1 - \nu_i^2)}{E_i} \quad (4)$$

Scratch-test analysis

Scratch-test method involves the sliding contact of an indenter on the worksurface and the resistance of this surface to the sliding motion. Indeed, the tip indenter slides on the worksurface with specific load, speed, and length (Fig. 3(d)). The in-situ normal and lateral forces are measured and the dynamic friction coefficient (μ_D) is calculated as the ratio between the lateral force and the normal force. In this study, the considered scratching length is 10 μm in order to work on elementary flax fibers and PP matrix separately. μ_D is evaluated in function of sliding speed and load.

Scale Effect on the Tribo-Mechanical Response of Natural Fiber Composites

Multiscale Mechanical Response of Natural Fiber Composites

As explained in section “Tribomechanical Tests Approach”, the mechanical characterization of NFC is realized on elementary fibers and polymer matrix separately. The mechanical response of flax fibers and PP matrix has been first performed by nanoindentation using the AFM method with diamond Berkovich indenter ($r = 40 \text{ nm}$) and an applied load of 500 μN (Chegdani *et al.*, 2017).

Fig. 5 presents the indentation traces and the corresponding elastic modulus values for flax fibers and PP matrix. Fig. 5(a) and (c) show that nanoindentation technique is able to target flax fibers and PP matrix separately. The elastic modulus of PP matrix is more affected by the contact depth than that of flax fibers. With the same applied load, flax fiber generates contact depths between 170 nm and 210 nm (Fig. 5(b)), while PP matrix generates contact depths between 70 nm and 160 nm (Fig. 5(d)). Consequently, the elastic modulus of PP matrix ($\sim 1\text{--}2 \text{ GPa}$) is higher than that of flax fibers ($\sim 0.7\text{--}1 \text{ GPa}$) at the same applied load.

To increase the geometrical contact scale, Fig. 6 shows the elastic modulus values obtained by nanoindentation using the RPM technique with the Hysitron TI-950 device (Chegdani *et al.*, 2018b). In this investigation, a higher tip indenter radius than that of AFM has been used ($r = 100 \text{ nm}$) and an applied load range from 100 μN to 500 μN has been considered to generate a large range of contact depth.

Fig. 6(a) reveals a different mechanical response of flax fibers comparing to that of AFM indentation of Fig. 5(b). Indeed, Fig. 6(a) shows an elastic modulus of flax fibers between 5 GPa and 22 GPa for a contact depth range from 25 nm to 230 nm. These elastic modulus values are largely higher than that obtained by AFM indentation. On the other hand, the mechanical response of PP matrix shown in Fig. 6(b) is not significantly different from that of AFM nanoindentation method shown in Fig. 5(d) at the same contact depth range. With a tip indenter radius of 100 nm, the elastic modulus of flax fibers becomes higher than that of PP matrix.

The investigation of the tip indenter radius (r) effect has been performed also with $r = 150 \text{ nm}$ (Chegdani *et al.*, 2019) and $r = 400 \text{ nm}$ (Chegdani *et al.*, 2017). Fig. 7 gives a comparison of the elastic modulus obtained with the different tip indenter radii for flax fibers and PP matrix for the same applied load ($F_{max} = 500 \mu\text{N}$).

The elastic modulus of flax fibers shows a significant increase when increasing the tip indenter radius, while the elastic modulus of PP matrix remains almost constant. This demonstrates the scale effect on the mechanical response of flax fiber induced by the geometrical contact scale. The multiscale mechanical behavior of flax fibers shown in this study is strongly related to the multiscale

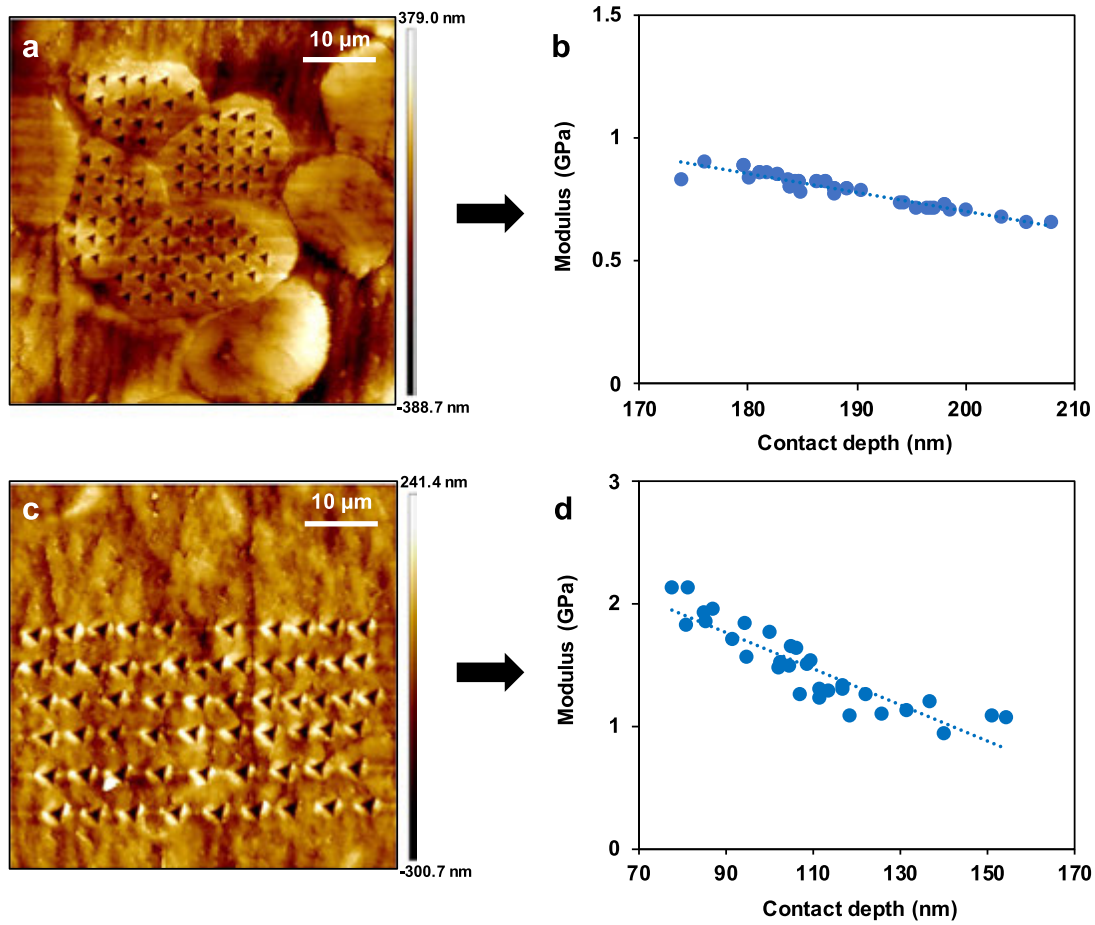


Fig. 5 (a) AFM scanning image of indented flax fibers. (b) Elastic modulus of flax fibers obtained by AFM nanoindentation. (c) AFM scanning image of indented PP matrix. (d) Elastic modulus of PP matrix obtained by AFM nanoindentation.

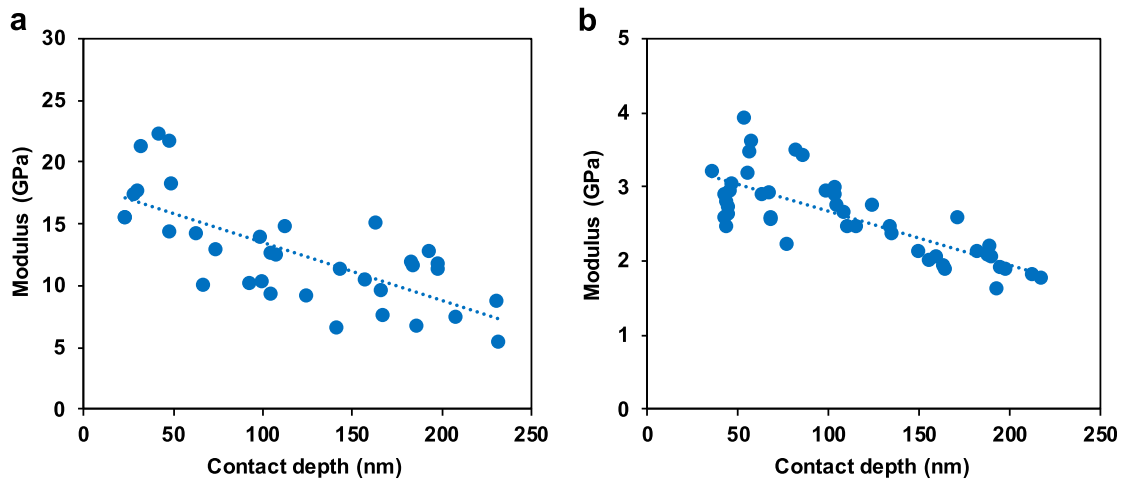


Fig. 6 (a) Elastic modulus of flax fibers obtained by RPM nanoindentation. (b) Elastic modulus of PP matrix obtained by RPM nanoindentation.

cellulosic structure of natural fibers (microfibrils → mesofibrils → elementary fibers) that affects the nature of the mechanical contact during the indentation. Indeed, when indenting with low tip indenter radius below the mesofibrils diameter (100–300 nm) (Bos *et al.*, 2004), the cellulose microfibrils (diameters between 1 and 4 nm (Bos *et al.*, 2004)) are transversally deviated from the indentation path as shown in Fig. 7(a). The mechanical response is almost that of non-cellulosic polymers in contact with the tip

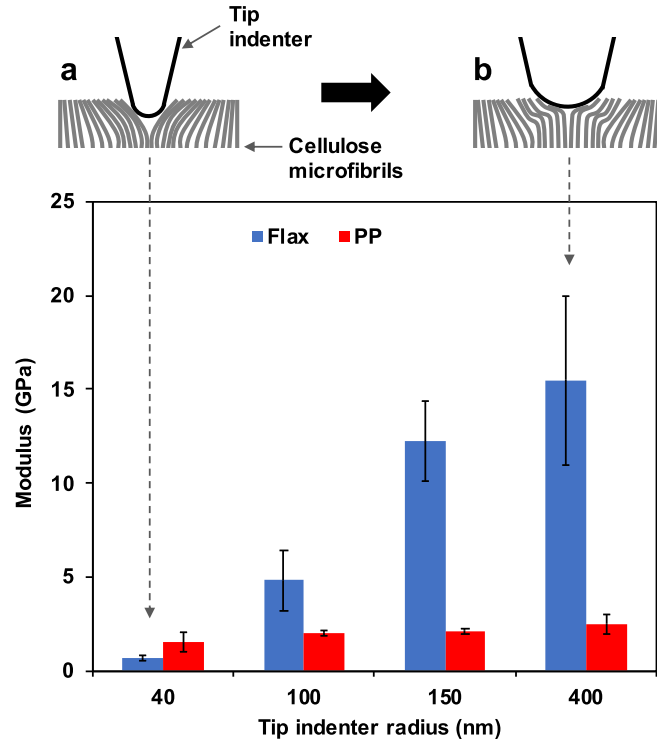


Fig. 7 Elastic modulus of flax fibers and PP matrix obtained with different tip indenter radii. (a) Schematic illustration of fiber/indenter contact with low indenter radius. (b) Schematic illustration of fiber/indenter contact with high indenter radius.

indenter. Since the tip indenter radius reaches the mesofibrils size, the indentation contact interface will include also the microfibrils that have high stiffness (135 GPa (Baley, 2002)) as shown in Fig. 7(b). Therefore, the indentation modulus increases by increasing the microfibrils contents in the contact area.

Multiscale Frictional Response of Natural Fiber Composites

The frictional response of NFC has been investigated by calculating the dynamic friction coefficient (μ_D) obtained from scratch-test experiments as the ratio between the scratching force (friction force) and the applied load (normal force). Scratch-test experiments were performed with AMF technique and RPM method by the Hisytron TI-950 device. Scratch-test experiments with AFM technique are not performed with the similar loading scale as for the RPM device. Indeed, lateral scratching motions with AFM indenters cannot be performed with high loads. Therefore, AFM scratch-tests were limited to an applied load of 30 μN . For the Hisytron TI-950 device, scratching experiments were performed with the Diamond indenter ($r = 100 \text{ nm}$) and Sapphire indenter ($r = 150 \text{ nm}$) with an applied load of 500 μN . Fig. 8 shows the typical scratching traces performed by RPM indenter. It can be seen that scratching traces of flax fibers and PP matrix have not the same shape. Plowed material is clearly observed at the bordered of each groove on PP matrix (Fig. 8(b)). For flax fibers, the grooves are formed without any noticeable plowing (Fig. 8(a)). The difference in the scratching mechanisms that occurs on flax fibers and PP matrix can be due to the high plasticity of PP matrix compared to flax fibers as shown in the nanoindentation study reported in section “Multiscale Mechanical Response of Natural Fiber Composites”.

Fig. 9 shows the dynamic friction coefficient (μ_D) generated by scratch-test experiments with AFM and RPM techniques. The first obvious observation is that AFM scratching friction coefficient (Fig. 9(a)) is not on the same order of magnitude than that of RPM scratching (Fig. 9(b)). Moreover, the friction coefficient of PP matrix is higher than that of flax fibers during the AFM scratching. However, the trend is reversed for the RPM scratching where the friction coefficient of flax fibers is higher than that of PP matrix. The reasons behind this difference could be related to both the contact and the load scales. Indeed, two scratching parameters are different between the two methods:

- The applied load: 30 μN for AFM scratching and 500 μN for RPM scratching.
- The tip indenter radius: 40 nm for AFM scratching and 100 nm for RPM scratching.

The variation of these two parameters affects the friction mechanisms. As well known, friction is a complex phenomenon that cannot be reduced to a single mechanism, but rather a result of a simultaneous action of various mechanisms at different hierarchy and scale levels such as adhesion, shear, and plowing (Chegdani and El Mansori, 2018; Nosonovsky and Bhushan, 2007). At low applied loads (30 μN), the predominant friction mechanism is adhesion that has an important role at nanoscale by controlling the

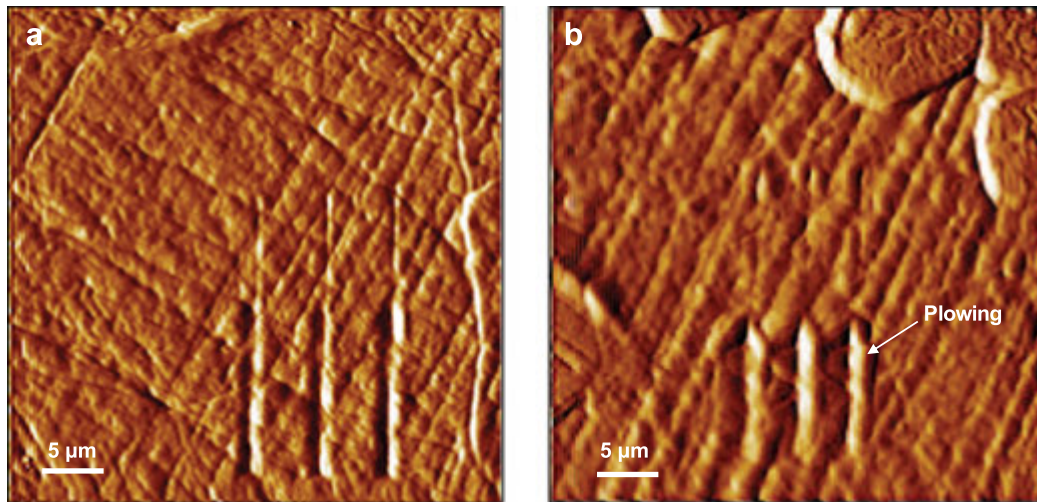


Fig. 8 Scratching grooves performed with RPM device using Diamond indenter at 500 μN of applied load on (a) flax fibers and (b) PP matrix.

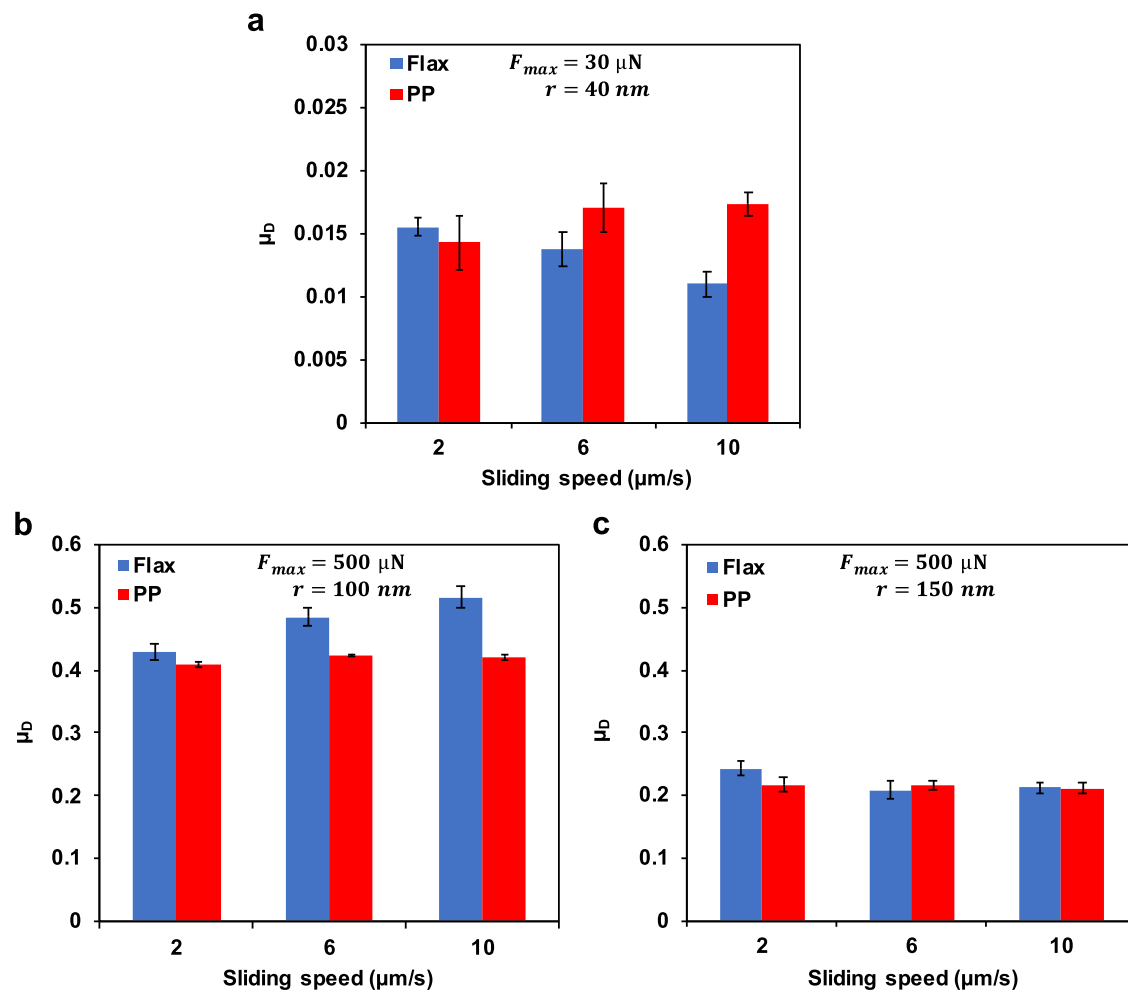


Fig. 9 Friction coefficient obtained by scratch-test with (a) AFM technique and diamond tip indenter, (b) RPM technique and diamond tip indenter, and (c) RPM technique and Sapphire tip indenter.

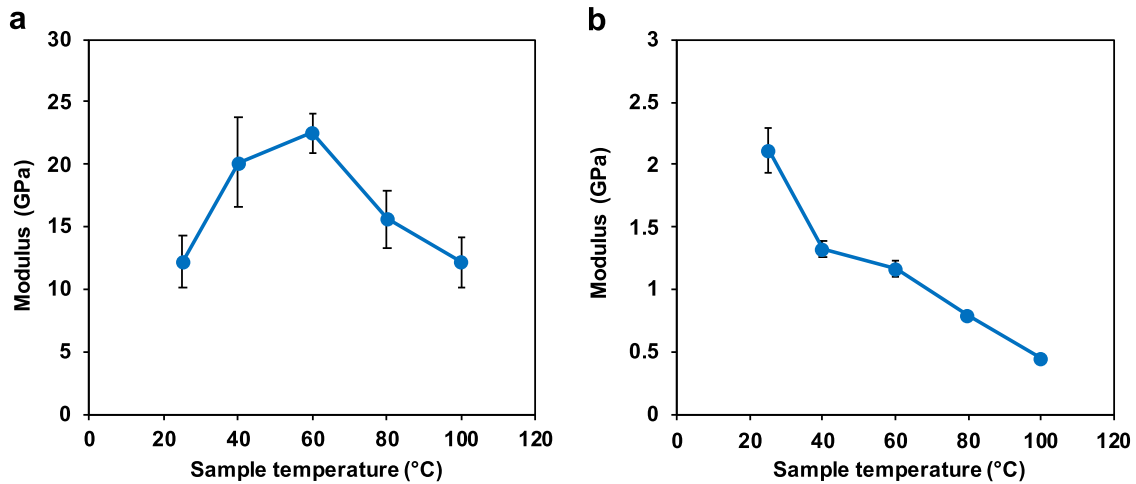


Fig. 10 Elastic modulus obtained by RPM nanoindentation at different sample temperatures for (a) flax fibers, and (b) PP matrix.

interaction forces between the atoms of the two surfaces in contact (Nosonovsky and Bhushan, 2007). This explains the low values of friction coefficient obtained with AFM scratching (Fig. 9(a)). Changing the loading scale ($30 \mu\text{N} \rightarrow 500 \mu\text{N}$) leads to the inclusion of further friction mechanisms such as shearing and plowing due to the increase of the contact depth of scratching. The friction coefficient is then upgraded to another scale level as shown in Fig. 9(b) for RPM scratching.

The frictional responses of flax fibers and PP matrix are also affected by the geometrical contact scale that controls the mechanical response as shown with nanoindentation experiments in section “Multiscale Mechanical Response of Natural Fiber Composites”. Indeed, the geometrical contact with an indenter radius of 40 nm engenders an elastic modulus of PP matrix higher than that of flax fibers. Consequently, the tangential scratching force of PP matrix is higher than that of flax fibers which is reflected on the friction coefficient results of Fig. 9(a). When upgrading the contact scale to 100 nm, the cellulose microfibrils come into play and increase the tangential scratching force of flax fibers which makes the friction coefficient of flax fibers higher than that of PP matrix as shown in Fig. 9(b).

On the other hand, the difference of RPM scratching friction between Diamond tip indenter (Fig. 9(b)) and Sapphire tip indenter (Fig. 9(c)) is mainly due to the effect of the tip material that leads to change the frictional properties between the tip and the worksurface.

Thermal Effect on the Tribo-Mechanical Response of Natural Fiber Composites

Thermomechanical Response of Natural Fiber Composites

The effect of sample temperature on the mechanical response of flax fibers and PP matrix has been performed with RPM nanoindentation technique using the Sapphire tip indenter (Chegdani *et al.*, 2019). A large sample temperature range has been considered from room temperature (25°C) to 100°C . Fig. 10 presents the evolution of the elastic modulus in function of sample temperature at iso applied load ($500 \mu\text{N}$) for flax fibers and PP matrix. Fig. 10(b) shows that increasing the sample temperature decreases significantly the stiffness of PP matrix. This reveals that the sample temperature affects strongly the softening of PP matrix. This thermo-mechanical behavior is well known in the literature for thermoplastic polymers (Drozdzov, 2010; Tripathi, 2002). Thermoplastic matrices soften under the effect of heat and become malleable at high temperatures with a significant decrease of the viscosity (Kannan *et al.*, 2013).

On the other side, the thermal effect on flax fibers is completely different as shown in Fig. 10(a). Indeed, in the temperature range of $[25\text{--}60^\circ\text{C}]$, the elastic modulus of flax fibers increases by temperature increase. However, in the temperature range of $[60\text{--}100^\circ\text{C}]$, the elastic modulus of flax fibers decreases by temperature increase. This specific behavior of flax fibers under thermal nanoindentation may be due to the chemical composition of their cellulosic structure shown in section “Multiscale Structure of Natural Fibrous Reinforcement in Composites”. Indeed, the cellulosic composition of natural fibers provides them a hydrophilic character that gives each elementary fiber the ability to absorb water molecules from the environment (Dittenber and GangaRao, 2012). Therefore, when indenting flax fibers from 25°C to 60°C , increasing the sample temperature leads to water release that acts as a plasticizer into the fiber structure (Hon and Shiraishi, 2000). This thermal effect on moisture content of flax fibers can explain the stiffness increase when heating flax fibers in the temperature range $[25\text{--}60^\circ\text{C}]$. Above 60°C , glass transition temperatures of the amorphous polymers inside the flax fiber are reached which makes the fiber softer. In fact, the glass transition temperature is 40°C for hemicelluloses, 50°C to 100°C for lignin, and above 100°C for cellulose (Kong *et al.*, 2017). Therefore, increasing the temperature above 60°C leads to exceeding the glass transition temperature of hemicellulose which is responsible of the bonding of cellulose microfibrils (Pere *et al.*, 2019). Consequently, the rigidity of fiber decreases when heating flax fibers in the temperature range $[60\text{--}100^\circ\text{C}]$.

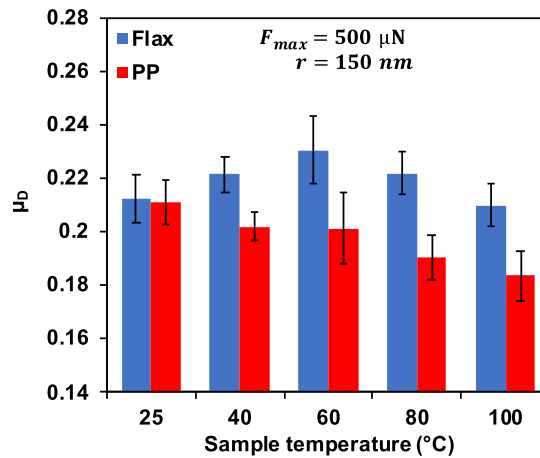


Fig. 11 Friction coefficient obtained by RPM scratch-test for flax fibers and PP matrix.

Thermo-Frictional Response of Natural Fiber Composites

Thermo-frictional analysis of NFC has been performed with scratch-test using the same experimental setup described in Section “Thermomechanical Response of Natural Fiber Composites” to reveal the effect of sample temperature. Fig. 11 presents the friction coefficient of flax fibers and PP matrix at different sample temperatures. The friction coefficient of PP matrix decreases by increasing the sample temperature. This frictional behavior at microscale is in good agreement with its mechanical response shown in Fig. 10(b) and also with the sliding friction of PP reported in literature at macroscale by standard tribometers where the sliding friction of PP polymer decreases with temperature increase (Ludema and Tabor, 1966). This is due to the polymer softening that leads to the decrease of the tangential scratching force (Chegdani *et al.*, 2019).

The frictional response of flax fibers shows a specific behavior that is similar to their mechanical behavior presented in section “Thermomechanical Response of Natural Fiber Composites”. Indeed, the friction coefficient of flax fibers increases by increasing the sample temperature from 25°C to 60°C and decreases by heating the samples even further from 60°C to 100°C as shown in Fig. 11. This demonstrated once again the local functional relationship between the mechanical response and the frictional response of each composite component. As for PP matrix, increasing the rigidity of flax fibers increases their tangential scratching force which contributes to a rise of the friction coefficient.

Conclusions

In this article, the tribo-mechanical behavior of natural fiber composites (NFC) has been investigated by performing nanoin-dentation and scratch-test experiments on flax fiber reinforced polypropylene matrix at different scale levels. The following conclusions can be drawn:

- Unlike PP matrix, the elastic modulus of flax fibers shows a tough dependence on the geometric contact scale where increasing the indenter tip radius increases significantly the fiber stiffness. Therefore, flax fibers engender multiscale mechanical properties that are related to their multiscale cellulosic structure.
- The local friction behavior of flax fibers and PP matrix shows a strong functional relationship with their respective local mechanical properties.
- The mechanical properties of NFC show a significant thermal effect that differs from flax fibers to the PP matrix. The thermal effect on mechanical properties is thus transposed to the frictional behavior of flax fibers and PP matrix.

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Mechanical Properties and Water Sorption of Chemically Modified Natural Fiber-Based Composites

Maya J John, Centre for Nanostructures and Advanced Materials, Council for Scientific and Industrial Research, Pretoria, South Africa; Department of Chemistry, Nelson Mandela University, Port Elizabeth, South Africa; and School of Mechanical, Industrial and Aeronautical Engineering, University of the Witwatersrand, Johannesburg, South Africa

Tshepiso P Molaba, Department of Chemistry, Nelson Mandela University, Port Elizabeth, South Africa and Polymers and Composites, Council for Scientific and Industrial Research, South Africa

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Introduction

The increasing use of glass fiber reinforced petroleum-based products has resulted in depletion of petroleum resources and entrapment of non-biodegradable materials in the environment. Biopolymers or synthetic polymers reinforced with natural fibers can be considered to be a viable alternative to glass fiber-based composites (John and Thomas, 2008). Natural fiber production depends mainly on solar energy and processing and extraction of fibers from plants utilizes small quantities of fossil fuel energy, while glass fiber production is an intensive process depending mainly on fossil fuels (Joshi *et al.*, 2004).

The growing interest in the use of natural fibers in composite applications is due to its many advantages compared to glass fibers such as low tool wear, low density and cost, world-wide availability and biodegradability (Ahmad *et al.*, 2015). The biodegradability of natural fibers can contribute to a healthy ecosystem while their low cost and high performance makes it suitable for use in industrial applications. The processing of natural fibers is more environmentally friendly, offering better working conditions and therefore reduces the risk of dermal or respiratory problems. The abrasive nature of natural fibers is much lower which leads to advantages with respect to both technical and recycling processes of composite materials. Automotive, motorsport and aerospace applications represent the best opportunity for application of natural fibers due to the favorable properties like lower weight, better crash absorbance and sound insulation properties. Recently, a survey regarding the possible applications of natural fibers in automobiles has been documented and presents that the use of natural fibers in advanced industrial sectors is on the increase.

There are however some disadvantages to using natural fibers, namely variable quality, lower impact strength, lower durability and moisture absorption (Araujo *et al.*, 2008; Celino *et al.*, 2014). Plant fibers, in particular, have hydroxyl groups which make them hydrophilic in nature and the absorption of moisture can affect composite properties such as mechanical performance (Celino *et al.*, 2013; Mokhothu and John, 2015). The presence of these hydrophilic groups at the fiber surface also does not favor compatibility between the fibers and typically hydrophobic polymer matrices, hence the fibers tend to form aggregates during composite processing and attain a poor degree of dispersion (Azwa *et al.*, 2013). It is therefore necessary in many instances to modify the fiber surface to render it more hydrophobic and also more compatible with resin matrices, and a number of fiber surface treatments have been employed by researchers to reduce moisture sensitivity of natural fibers (Aseer *et al.*, 2015).

Natural fibers can be sourced from plants or animals. Hence, these fibers are in abundance around the world. Depending on their origin, natural plant fibers may be grouped into: bast (stem), leaf and seed types. The best known examples are: (1) Bast: jute, flax, kenaf, hemp and ramie; (2) Leaf: sisal, banana and pineapple leaf fiber (PALF); (3) Seed/fruit: cotton, coir and kapok (Namvar *et al.*, 2014). The chemical composition of plant fibers varies depending upon the type of fiber. Such variations may also be influenced by the origin, age and retting (mode of extraction of fiber from the source) process adopted. The chemical composition as well as the structure of fibers is fairly complex (Jamrichova and Akova, 2013).

The following sections gives an overview on the different types of chemical modifications and cites important studies that highlight results of improved mechanical and water sorption properties in chemically modified natural fiber composites.

Chemical Composition of Plant Fibers

Plant fibers are by far the most used in natural fiber reinforced composites. As stated already, moisture absorption can be a disadvantage. The main constituents in natural fibers are composed of cellulose, hemicellulose, lignin, pectins and waxes (Nguong *et al.*, 2013). Moisture absorption is related to the chemical composition of the fiber. Cellulose is a polymer of β -D-Glucose, which in contrast to starch, is oriented with $-\text{CH}_2\text{OH}$ groups alternating above and below the plane of the molecule thus producing long, unbranched chains (Fig. 1). The absence of side chains allows cellulose molecules to lie close together and form rigid structures. Cellulose is the major structural material of the fiber. The large amount of hydroxyl groups in cellulose gives natural fibers their hydrophilic properties which, when used to reinforce hydrophobic matrices, results in a very poor interface and poor resistance to moisture absorption. The mechanical properties of natural fibers depend on the type of cellulose present. This is because each type of cellulose has its own cell geometry and geometrical conditions determine the mechanical properties. Solid cellulose forms a microcrystalline structure with regions of high order, i.e., cellulose regions. There are several different crystalline arrangements of cellulose. These polymorphs of cellulose are denoted cellulose I, II, III_I, III_{II}, IV_I, and IV_{II}. They can be interconverted depending on chemical treatment and source. For a long time, native cellulose attracted the interest of a large scientific community in an attempt to elucidate its crystal structure. Native cellulose, namely cellulose I, is crystalline cellulose and it does not contain intersheet hydrogen bonding. Cellulose II, also called

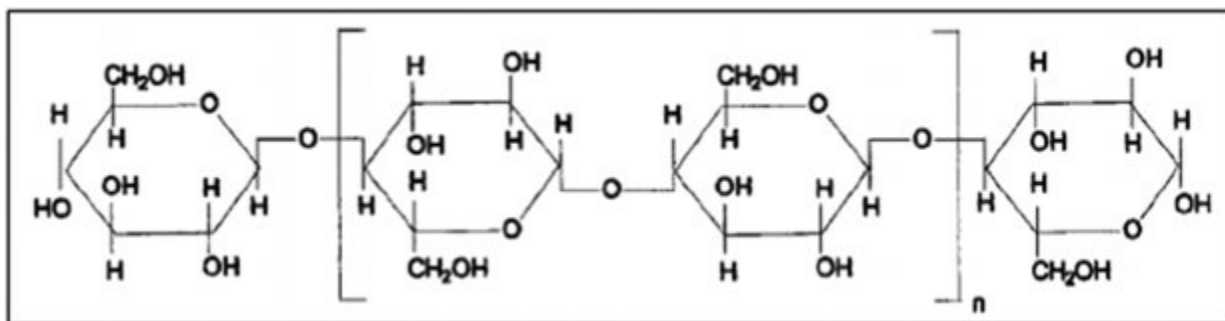


Fig. 1 Molecular structure of cellulose. Reproduced from Mohanty, A., Misra, M., Drzal, L., 2001. Surface modifications of natural fibres and performance of the resulting biocomposites: An overview. *Composite Interfaces* 8, 313–343.

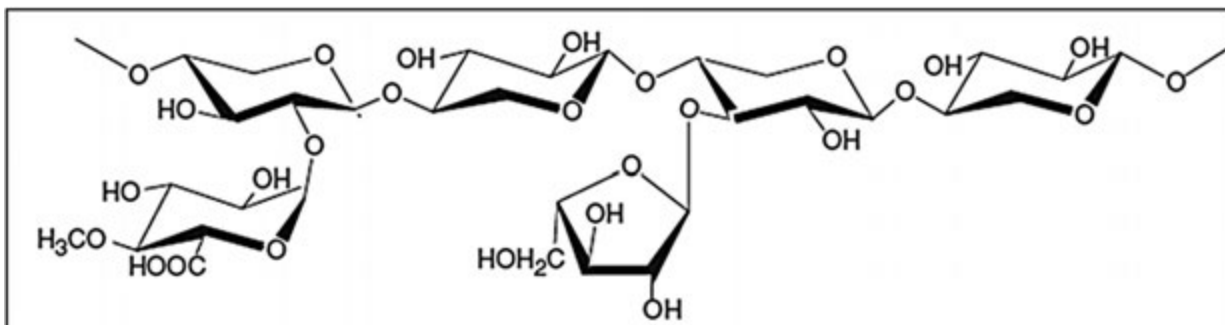


Fig. 2 Molecular structure of hemicellulose (Downloaded from internet: Biomass Conversion and Composition).

generated cellulose, is cellulose precipitated out of solutions and it contains intersheet hydrogen bonding. Cellulose I and II represent the two main polymorphs of cellulose (Siqueira *et al.*, 2010). Cellulose I is not the most stable form of cellulose and is responsible for mechanical properties due to its high modulus and crystallinity. An additional hydrogen bond per glucose residue in cellulose II makes this allomorph the most thermodynamically stable form (Saxena and Brown, 2005).

Hemicellulosic polymers are branched, fully amorphous and have significantly lower molecular weight than cellulose (Prasad *et al.*, 2013). The chemical structure of hemicellulose consists of long chains of a variety of pentoses, hexoses and their corresponding uronic acids. The open structure containing many hydroxyl groups and acetyl groups makes it partly soluble in water and hygroscopic. Hemicellulose is strongly bound to cellulose fibrils presumably by hydrogen bonds (Fig. 2). Unlike cellulose, the constituents of hemicellulose differ from plant to plant. The degree of polymerization (DP) of hemicellulose is around 50–300, and of native cellulose is 10–100 times higher than that of hemicellulose (Hallac and Ragauskas, 2011).

The exact chemical nature of lignin still remains obscure. The main difficulty in lignin chemistry is that no method has so far been established by which it is possible to isolate the lignin from the native state from the fiber (Mohanty *et al.*, 2002). Although the structural formula of lignin in natural fiber has yet not been established, most of the functional groups that make up the molecule have been identified. The proposed model for lignin is shown in Fig. 3. Lignin is amorphous, highly complex and mainly aromatic. Hydroxyl, methoxyl and carbonyl groups have been identified. Lignin has been found to contain five hydroxyl and five methoxyl groups per building unit. It is believed that the structural unit of the lignin molecule are 4-hydroxy-3-methoxy phenylpropane derivatives (Mishra *et al.*, 2004). Lignin has the least water sorption of all the natural fiber components. It gives rigidity to the plants. During synthesis of plant cell walls, polysaccharides such as cellulose and hemicellulose are laid down first and lignin fibrils fills the spaces between the polysaccharides fibers, cementing them together. This lignification process causes stiffening of cell walls, and the carbohydrate is protected from chemical and physical damage. It is not hydrolyzed by acids, but soluble in hot alkali, readily oxidized and easily condensable with phenol. The mechanism of lignin biosynthesis is still under active research.

Pectins are a collective name for heteropolysaccharides which consists of polygalacturon acid. Pectin is soluble in water only after a partial neutralization with alkali or ammonium hydroxide. They give plants flexibility. Waxes are also a constituent of fibers, which can be extracted with organic solutions. These waxy materials consist of different types of alcohols, which are insoluble in water as well as in several acids (palmitic acid, oleaginous acid, stearic acid).

Moisture Absorption in Natural Plant Fiber Reinforced Composites

Natural plant fibers, being hydrophilic in nature, absorb moisture from the environment. Hydrogen bonds are formed between the –OH groups of the cellulose molecules in natural fibers and water. Moisture content in natural fibers (NF) reaches around

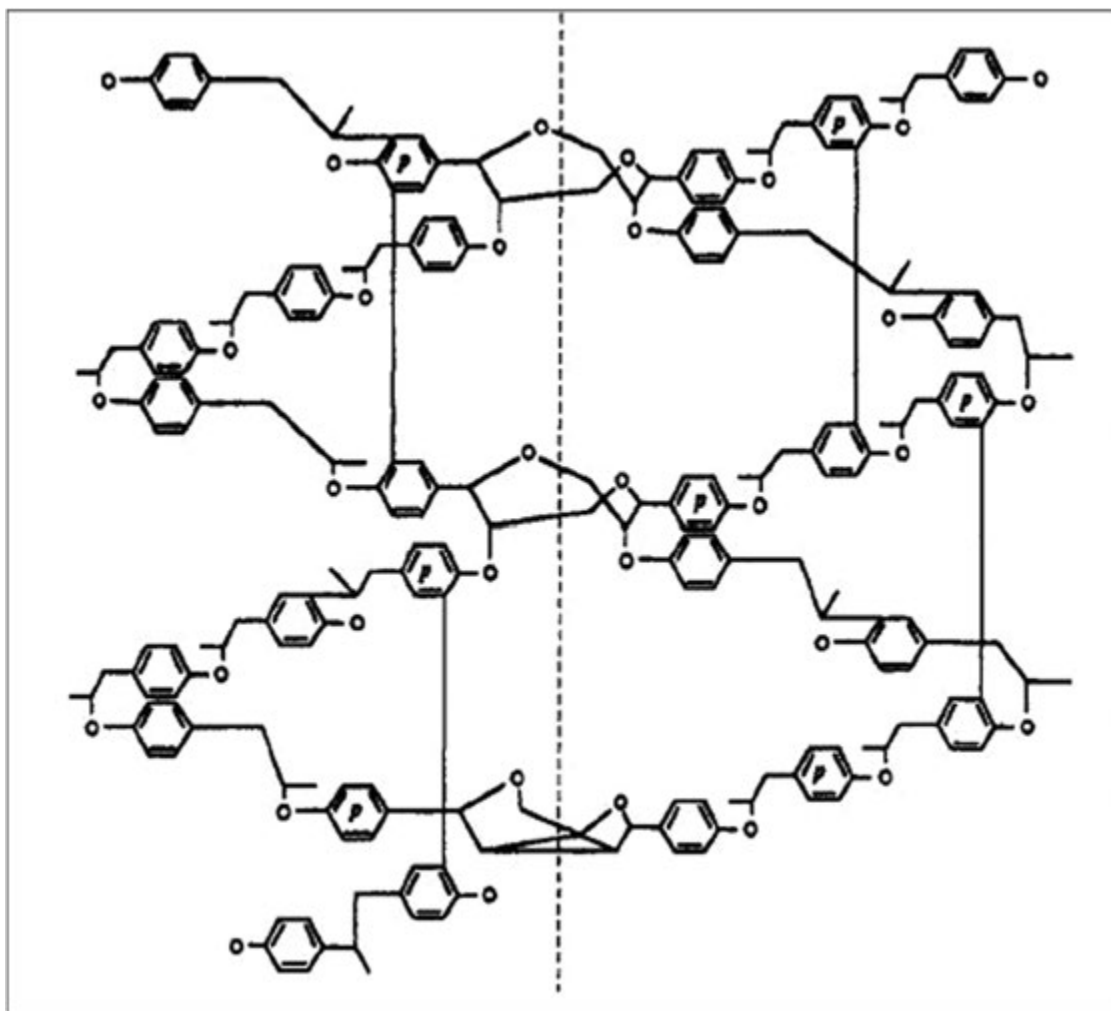


Fig. 3 Proposed model for lignin. Reproduced from Faulon, J.-L., Hatcher, P.G., 1994. Is there any order in the structure of lignin?. *Energy & Fuels* 8, 402–407.

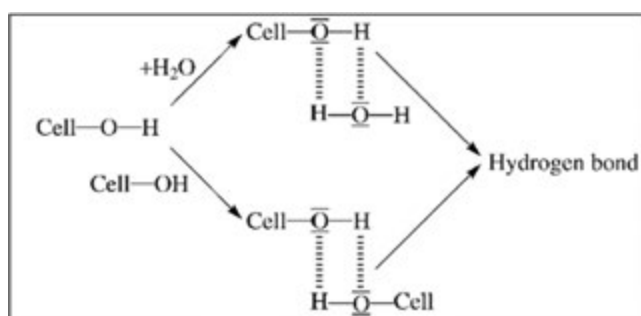


Fig. 4 Schematic representation of the interaction of cellulose with water molecules. Reproduced from Jacob, M., Varughese, K., Thomas, S., 2005. Water sorption studies of hybrid biofibre-reinforced natural rubber biocomposites. *Biomacromolecules* 6, 2969–2979.

8%–12.6%. The first step in H_2O sorption is the absorption of water onto $-\text{OH}$ groups in natural fiber. The second step is attachment of water molecules either to other hydrophilic groups of which they may form further layers on top of the water molecules already absorbed. The interaction of cellulose with water molecules is shown in Fig. 4. Thus, these natural fibers can absorb and desorb moisture from the atmosphere and equilibrate with the surrounding environment. Hence composites reinforced with such fibers undergo swelling and shrinkage in moist and dry environments, respectively, affecting their dimensional stability. Drying of the fibers before processing is an important requirement because water on the fiber surface acts like a separating

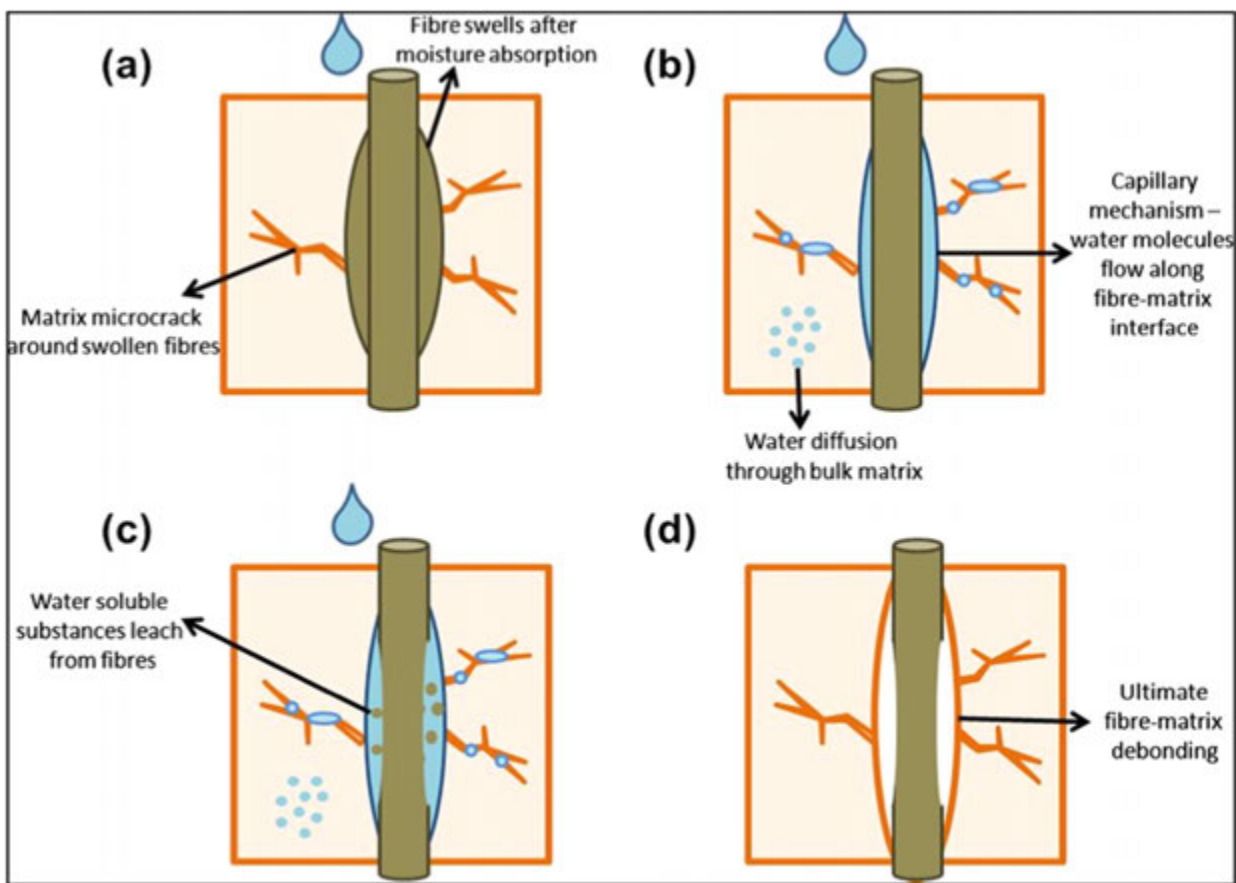


Fig. 5 Effect of water absorption on fiber matrix interface. Reproduced from Azwa, Z., Yousif, B., Manalo, A., Karunasena, W., 2013. A review on the degradability of polymeric composites based on natural fibres. *Materials & Design* 47, 424–442.

agent in the fiber-matrix interface. Water transport is strongly affected by the nature of the fiber-matrix interface. If the interface is strong it is difficult for the water molecules to diffuse into the composite system. Upon various chemical treatments (alkali, silane treatment) of the fiber, one can achieve strong interfacial interaction, thereby making the composite more resistant to water transport.

Mechanical properties of fiber-reinforced composites depend on the nature of the polymer matrix, distribution and orientation of the reinforcing fibers, the nature of the fiber-matrix interfaces and of interphase region. The mechanical properties of natural fiber reinforced composites decrease considerably when they are exposed to moisture. Besides affecting the properties of the polymer matrix and natural fibers themselves, moisture can seriously damage the fiber-matrix interface leading to poor stress transfer efficiencies. The degradation process starts with swelling of cellulose fibers that develops stress at the interface and causes micro-cracking of the matrix around the swollen fibers. The absorbed water starts to establish intermolecular hydrogen bonding with fibers and thereby reduces the interfacial adhesion between the fiber and the matrix and water soluble substances start leaching out from the fibers. This eventually leads to ultimate debonding between fiber and the matrix. This process is summarized in (Fig. 5).

It has also been reported that the water absorption of cellulosic fiber composites depends on the fiber content and temperature. As the temperature of absorption of water increases, more water penetrates into the composites and swells up the fibers causing a reduction in tensile strength.

Mechanical and Water Sorption Studies in Chemically Treated Natural Fibers and Composites

A number of fiber surface treatments have been employed to reduce moisture sensitivity of natural fiber reinforced composites. The natural fiber surface can be modified by physical treatments (cold plasma treatment, corona treatments) and chemical treatments (coupling agents, alkali, maleic anhydride, organosilanes, isocyanates, sodium hydroxide, permanganate and peroxide). Chemical modification is imparted to improve adhesion between the hydrophilic natural fiber and hydrophobic polymer matrix and also to decrease moisture absorption leading to the improvement of mechanical properties.

The chemicals used for surface modification must be capable of reacting with lignocellulosic hydroxyls under neutral, mild alkaline or acidic conditions at temperatures below 150°C (Bogoeva-Gaceva *et al.*, 2007). The chemical system should be simple and capable of swelling the structure to facilitate penetration. The treated fiber must still possess the desirable properties of untreated fibers. Mechanical properties of the fiber should not be reduced, good electrical insulation properties should be retained and the hydrophobic nature of reagent should be selected accordingly.

The common chemical modification techniques are the following:

Alkali Treatment

Alkali treatment results in an increase in the amount of amorphous cellulose at the expense of crystalline cellulose. The important modification occurring here is the removal of hydrogen bonding in the network structure. The effect of alkali on a cellulose fiber is a swelling reaction, during which the natural crystalline structure of the cellulose relaxes. The type of alkali (KOH, LiOH, NaOH) and its concentration will influence the degree of swelling, and hence the degree of lattice transformation into cellulose-II. Studies have shown that Na⁺ has got a favorable diameter, able to widen the smallest pores in between the lattice planes and penetrate into them. Consequently, sodium hydroxide treatment results in a higher amount of swelling. This leads to the formation of new Na-cellulose-I lattice, a lattice with relatively large distances between the cellulose molecules, and these spaces are filled with water molecules. In this structure, the OH-groups of the cellulose are converted into ONa-groups, expanding the dimensions of molecules. Subsequent rinsing with water will remove the linked Na-ions and convert the cellulose to a new crystalline structure, i.e., cellulose-II, which is thermodynamically more stable than cellulose-I. Sodium hydroxide can cause a complete lattice transformation from cellulose-I to cellulose-II, in contrast to other alkalis that produce only partial lattice transformation. The alkali solution influences not only the cellulosic components and the non-cellulosic components such as hemicellulose which is removed by the action of alkali.

Acetylation

Acetylation is a rather attractive method of modifying the surface of natural fibers and making it more hydrophobic. It has been shown to reduce swelling of wood in water and has been studied more than any other chemical reaction of lignocellulosic materials. The main principle of the method is to react the hydroxyl groups (OH) of the fiber with acetyl groups (CH₃CO), therefore rendering the fiber surface more hydrophobic. The hydroxyl groups that react are those of the minor constituents of the fiber, i.e. lignin and hemicelluloses, and those of amorphous cellulose. The hydroxyl groups in the crystalline regions of the fiber are closely packed with strong interchain bonding, and are inaccessible to chemical reagents. Acetylation has been shown to be beneficial in reducing moisture absorption of natural fibers. Reduction of about 50% of moisture uptake for acetylated jute fibers and of up to 65% for acetylated pine fibers has been reported in the literature (Bledzki and Gassan, 1999). Acetylation has also been found to enhance the interface in flax/polypropylene composites.

Silane Treatment

These chemicals are hydrophilic compounds with different groups appended to silicon such that one end will interact with matrix and the other end can react with hydrophilic fiber which act as a bridge between them. The uptake of silane is very much dependent on a number of factors including hydrolysis time, organofunctionality of silane, temperature and pH. Alkoxy silanes are able to form bonds with hydroxyl groups. Silanes undergo hydrolysis, condensation and the bond formation stage. Silanols can form polysiloxane structures by reaction with hydroxyl group of the fibers. The chemical reaction is given in Fig. 3.

In addition to the self-condensation of silanes and the condensation on the surface of cellulose fiber, amino silane molecules can interact with the OH groups of cellulose via their Brønsted basic amino groups. Studies (FTIR and XPS spectroscopy) have also shown that the reaction between silanes and cellulose takes place only at temperatures above 70°C (Valdez Gonzalez *et al.*, 1999).

The influence of different types of chemical modifications: NaOH, peroxide, permanganate and stearic acid on Agave americana fibers and the effects on the chemical composition, structural characteristics, crystallinity, thermal degradation, tensile properties and surface morphology were studied by Madhu *et al.* (2020). The authors observed that the average tensile strength and elongation at break of chemically treated fibers are higher than those of raw fibers and this was attributed to the fiber structure becoming closely packed due to the removal of hemicellulose by various chemical treatments.

Osman *et al.* studied water absorption and mechanical properties of kenaf-recycled jute fibers reinforced unsaturated polyester composites (Osman *et al.*, 2012). Kenaf and recycled jute fibers were subjected to NaOH treatment. Composites were immersed in distilled water at room temperature. Flexural properties of kenaf fiber composites decreased drastically on exposure to water immersion. This was due to the formation of hydrogen bonding between the water molecules and cellulose fiber. Alternatively, incorporated recycled jute fiber in composites decreased water uptake and enhanced dimensional stability. The reason for that was the percentage of holocellulose for the jute is 87.6% which is greater than holocellulose of kenaf 62%, and this led to the improvement of flexural properties of hybrid composites.

Robertson *et al.* investigated the mechanical performance and moisture absorption of various natural fiber reinforced thermoplastic composites (Robertson *et al.*, 2013). Natural fiber composites made from low density polyethylene (LDPE) and a variety

of Canadian based fiber feedstocks (hemp, flax, chemically pulped wood, wood chips, wheat straw and mechanically pulped triticale) were examined. All natural fibers tested exhibited various levels of water uptake over a period of 11,515 h (68 weeks or approximately 16 months). Increase in fiber content resulted in an increase in water absorption. At 10 wt% fiber fraction, all fiber types absorbed similar amounts of water (approximately 2% mass gain in 16 months), while at other fiber fractions, there was a significant difference between fiber types. At 40 wt% fiber fraction, the pulped wood composite had the lowest overall moisture gain of 6.3%, while other fiber types absorbed between 11.1% and 13.5% water. This was attributed to the observed difference in fiber geometries and size distribution. Addition of maleic anhydride polyethylene (MAPE) resulted in a decrease in water uptake for all fiber types. The most significant impact was seen for hemp, flax, wood chips and wheat straw. The increase in bonding between the fiber and matrix (as a result of adding of MAPE) reduced moisture sorption in these composites.

Chen *et al.* studied the effects of interfacial modification and high fiber loading on rice husk flour (RHF) fibers and recycled polymer blend (Chen *et al.*, 2015). Fiber surface treatments such as NaOH and maleic anhydride polyethylene (MAPE) were carried out. The composites modified with only MAPE exhibited lowest water absorption. The compatibility between the hydrophilic rice husk fiber and the hydrophobic recycled polymer blend matrix was enhanced by incorporating MAPE because of the esterification reaction that occurred between the anhydride moieties in MAPE and the surface hydroxyl groups of the rice husk flour fibers. Upon esterification, the ethylene groups of MAPE attaches to the fiber surface, blocking pores and lumen; thus water molecules were prevented from being absorbed.

Doroudgarian *et al.* studied moisture uptake and resulting mechanical response of bio-based composites (Doroudgarian *et al.*, 2014). Composites were conditioned in desiccators (until moisture level content reached saturation) at different relative humidity (RH) levels: 41%, 70% and 98%. Flax fibers and regenerated cellulose fiber rovings were subjected to chemical treatments such as NaOH and 3-aminopropyltriethoxy silane. Stiffness and strength of the composites were drastically reduced by moisture at RH = 98%, whereas the strain was increased. The fiber treatment did not improve resistance of cellulose fibers to moisture. According to the authors, treatment was limited to the surface of the fibers without significant penetration. If the chemical treatment would be able to affect larger volume of fiber (throughout the fiber) a greater protection against moisture could be achieved.

Zhu *et al.* studied the effect of fiber treatments on water absorption and tensile properties of flax/tannin composites (Zhu *et al.*, 2013). Water absorption tests were conducted by immersing the specimens in distilled water at room temperature. Chemical treatments such as NaOH, butanetetracarboxylic acid (BTCA), aminopropyltriethoxy silane (APS), benzenediol (Laccase) and dodecyl gallate (Laccase-Doga) were applied on nonwoven flax fibers. The water sensitivity of composites was not reduced by these treatments as expected. The decrease in tensile strength was about 20% during 3 days of immersion for untreated composites. The value of NaOH and NaOH-BTCS treated composites dropped by 13% and 16%, respectively. For Laccase-Doga and Laccase treated composites the strength dropped by 28% and 35%. The Young's modulus of neat composites decreased about 45% at saturation, while the reduction of young's modulus for treated composites was also over 40%.

Abdullah *et al.* studied the effect of chemical treatment on mechanical and water-sorption properties of coconut fiber reinforced unsaturated polyester (recycled PET) composites (Munirah Abdullah and Ahmad, 2012). Composites were immersed in distilled water at room temperature. Chemical treatments such as alkali and silane were applied. Treated composites showed lower water absorption than untreated composites. Silane treated composites showed lesser water absorption in all sample. The order of decreasing value of water absorption composites was as follows: untreated < silane < alkali < silane on alkali treated coconut fibers. Good fiber/matrix adhesion upon chemical treatment was found to be the main reason for the decreased water uptake.

Ali *et al.* studied the effect of single and double stage chemically treated kenaf fibers on mechanical properties of kenaf fiber reinforced polyvinyl alcohol (PVA) biocomposites films (Ali *et al.*, 2014). Composites were exposed in the environmental chamber at 75% RH and 22°C. Kenaf fibers were treated with chemical treatments such as NaOH, single stage treatment (SST) and double stage treatment (DST). Single stage treatment involved treatment of the fibers with chromium (III) salt solution for 3 h. Double stage treatment was performed using the same procedure as the single stage treatment with an additional 2 h in a solution of chromium (III) salt solution. The moisture uptake (%) increased with the increase of fiber content and duration of exposure days for all the treated and untreated kenaf fiber reinforced PVA composites. Composite reinforced with DST kenaf fiber exhibited the lowest moisture uptake properties, followed by composites reinforced with SST, alkali treated and raw kenaf. This was attributed to the decrease of the hydroxyl groups as well as the micro-voids in the composites responsible for moisture absorption. During SST and DST, the chromium (III) ions formed complexes with cellulose hydroxyl groups, which would decrease the relative abundance of free hydroxyl groups for the formation on inter and/or intra molecular hydrogen bonds in the cellulose molecules of kenaf fiber. This resulted in a decrease of fiber agglomeration and improved fiber dispersion within PVA matrix, which reduced microvoids.

Zabihzadeh investigated water uptake and flexural properties of wheat-straw, pine and poplar reinforced polyethylene composites (Zabihzadeh, 2009). Water absorption was determined after 2 h and 24 h immersion in distilled water at 30°C, 45°C, 60°C and 75°C. Wheat straw composites absorbed more water compared to poplar and pine composites. This was attributed to the presence of a high amount of pentosans and low amount of lignin in wheat straw filler. Cellulose and hemicellulose are mostly responsible for the high water absorption of natural fibers, since they contain numerous hydroxyl groups while lignin is a hydrophobic material. The composites with 2% compatibilizer (MAPE) showed lower water absorption compared to those without compatibilizers. The extent of water absorption of compatibilized samples was about 25%, 15% and 26% less than that of uncompatibilized wheat straw, poplar, and pine composites, respectively for 2 h immersion time. The anhydride groups present in MAPE can covalently bond to the hydroxyl groups of the fiber surface. The decrease of water absorption was therefore attributed to the occupation of free hydroxyl groups.

Sampathkumar *et al.* studied the effect of chemical treatment on water absorption of areca fiber. Areca fibers treated with 15% NaOH shown maximum water uptake in pond water and river water and fiber treated with 5% NaOH showed maximum water absorption in sea water (Sampathkumar *et al.*, 2012). The authors suggested that the swelling process was much faster leading to higher water uptake and also the alkali treatment had removed waxy substances and natural oils covering the cell wall of the fibers thus increasing water absorption. In another study by Mohd Nazarudin *et al.* alkali treatment resulted in lower water uptake of non-woven kenaf fiber reinforced polyester composites (Mohd Nazarudin *et al.*, 2013).

Dixit *et al.* studied the effect of surface modification on the water absorption behavior of coir fibers (Dixit and Verma, 2012). Coir fibers were subjected to chemical treatments such as alkali, acetylation and permanganate treatment. Water absorption characteristics of treated coir were studied by immersion in distilled water at room temperature and at 50°C for 24 h. They reported that acetylation and permanganate treated coir showed significant decrease in water absorption capacity of coir. But the most successful treatment was acetylation which showed up to 36% decrease at room temperature and 47% at 50°C respectively in moisture absorption tendency of coir fibers. This appeared to be due to the fact that chemical treatment successfully modified the structure of coir fibers by bonding between $-CH_3CO$ groups and $-OH$ groups of coir fibers.

Tajvidi *et al.* studied long term water uptake behavior of kenaf and rice hulls reinforced PP composites (Tajvidi *et al.*, 2006). Water absorption behavior of the composites was studied and water diffusion coefficients were also calculated by evaluating the water absorption isotherms. They found that water absorption was affected by fiber type. Kenaf fiber/PP composites exhibited the highest water absorption while the lowest water absorption was observed for rice hulls/PP composites. This behavior was attributed to high amount of extractives and ash and lower amount of cellulose and pentose present in rice hulls. Higher fiber content resulted in higher water absorption. The effect of fiber content on water absorption was more pronounced at higher soaking time. At higher fiber contents, water uptake in composites could not reach to saturation point within five weeks immersion in water. Dorgan *et al.* studied the water absorption characteristics of kenaf fiber reinforced PLA composites (Munirah Abdullah and Ahmad, 2012). They reported that swelling and water absorption of the biocomposites increased with increasing kenaf fiber content and their values were very high when compared to pure PLA. Treatment of the biocomposites containing 50% fiber with silane coupling agent reduced swelling and water absorption compared to the untreated biocomposites.

Abdelmouleh *et al.* studied the effects of coupling agent and fiber loading on short natural-fiber reinforced polyethylene and natural rubber composites (Abdelmouleh *et al.*, 2007). Fibers were submitted to chemical treatments such as γ -methacryloxypropyltrimethoxy (MPS), γ -mercaptopropyltrimethoxy (MRPS) and hexadecyltrimethoxyl-silanes (HDS). Samples were weighed and then soaked in distilled water at 25°C. Four different cellulose fibers with different average lengths were used, namely Avicel, Technocel – 2500, Alfa pulps and Pine fibers. For composites containing 50 wt% alfa fibers subjected to different fiber surface treatments, water absorption was found to increase with immersion time reaching a plateau level after about 6 days. The equilibrium water uptake was found to depend on the treatment of the fibers. LDPE composites containing untreated fibers absorbed 3.5% of water whereas the composites based on HDS, MPS and MRPS treated fibers absorbed 2.2%, 2.6% and 3% of water.

Punyamurthy *et al.* studied the effect of alkali treatment on water absorption of single cellulosic abaca fiber (Punyamurthy *et al.*, 2012). The comparison of moisture absorption studies of untreated and alkali-treated abaca fiber in sea water, pond water, river water and borewell water revealed that the moisture absorption capability of abaca fiber was reduced by alkali treatment. Also the moisture absorption of the treated fiber decreased with an increase in alkali concentration. Alkali treated fiber absorbed less moisture in pond water (pH = 8.2) than in the other water samples. Abaca fiber treated with 20% alkali showed about 38.27, 52.94, 62.50 and 63.50% lower moisture absorption properties than the untreated abaca fiber in river water, borewell water, sea water and pond water, respectively, after immersing the fiber in a water sample for about 624 h.

Vilay *et al.* studied the effect of fiber surface treatment (acrylic acid and sodium hydroxide) and fiber loading on the properties of bagasse fiber reinforced unsaturated polyester composites (Vilay *et al.*, 2008). They reported that the treated composites showed lower water absorption than the polyester resin. This was attributed to the acrylic acid and NaOH treatment of the bagasse fibers. From the diffusion coefficient (D) values of the composite samples, it was found that the NaOH and acrylic acid (AA) treated fiber polyester composites had better resistance towards water absorption than those of untreated fiber composites. Untreated fiber composites showed the highest D values. Higher D value might also indicate higher void content in the system where voids generate more pathways for water to start diffusing into the composite. Moisture content at saturation level values were found to be 12.18% for untreated composite, 8.08% for NaOH treated composite and 7.09% for acrylic acid treated composites.

John *et al.* studied effect of chemical modification on properties of hybrid sisal-oil palm reinforced composites (John *et al.*, 2008). Alkali treatment resulted in lowered water uptake in hybrid sisal-oil palm reinforced composites compared to the untreated samples. Among the chemically treated composites, composites containing fibers treated with 0.5% NaOH showed the highest water uptake while the composites containing fibers treated with 4% NaOH exhibited minimum water uptake. As the NaOH concentration increases, the adhesion between the fiber and matrix increases, and hence, uptake of water decreases. Also increased alkali concentration induced greater crystallinity of the fibers, thereby reducing the water sorption capability of the fibers.

Studies have also explored the use of plant-based coupling agents for improving mechanical performance in composites. These agents include utilizing zein (John and Anandjiwala, 2009; Whitacre *et al.*, 2015) (amphiphilic coupling agents), furfuryl alcohol (furanic monomers) and fungi (Gulati and Sain, 2006) for treating plant fibers. Fig. 6 represents the effect of furfuryl alcohol treatments on the water uptake of flax reinforced polyfurfuryl alcohol composites and shows the amount of water uptake after 243 h was reduced by 42.4%, compared with untreated composites. This was attributed to increased hydrophobicity of flax fibers after FA treatment and better fiber-matrix bonding of the composites which impede the propagation of water molecules along with

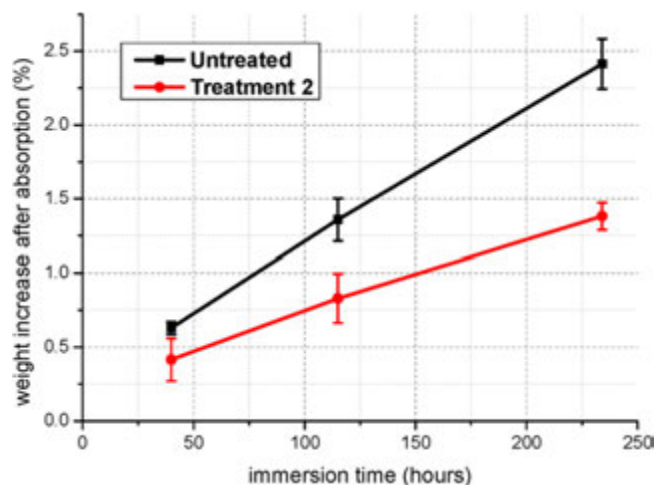


Fig. 6 Influence of FA treatments on the water uptake of flax-bio-based epoxy composites. Reproduced from John, M.J., Anandjiwala, R.D., 2009. Chemical modification of flax reinforced polypropylene composites. *Composites Part A Applied Science and Manufacturing* 40, 442–448.

the fiber-matrix interfaces (Jia and Fiedler, 2018). Other studies (Liu *et al.*, 2020) have investigated the deposition of nano-SiO₂ on jute fibers and observed an increase of tensile strength by 16.47% as compared to that of the control jute fibers. This was attributed to formation of a protective layer of nano-SiO₂ on jute fibers along with bonding between carbon and silicon groups.

It is clear from the above studies that in composites chemical treatments are most promising when there is effective covalent bonding between –OH groups of natural fiber and functional groups of coupling agent and this results in reduced water sorption and improved mechanical performance. It can therefore be concluded that there are different parameters that need to be taken into account when doing chemical modification of natural fibers such as:

- (1) Different concentrations of the reagents used
- (2) Time of treatment
- (3) Temperature
- (4) Type of fiber (woven, non-woven, short fibers)

Conclusion

This article covers the mechanical and moisture absorption studies in chemically treated plant fiber composites. Natural fibers and its composites are sensitive to moisture and the absorption mechanism in composite materials is mainly governed by diffusion of water molecules between polymer chains. It is evident that chemical modification can improve mechanical properties and water resistance in natural fiber reinforced composites. Careful selection of chemical modification techniques is thus essential because a balance must be met between maintaining the cost of the chemical agents and improving mechanical performance in composites.

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Machining Behavior of Natural Fiber Composites

Faissal Chegdani, Arts et Metiers Institute of Technology, Mechanics Surfaces and Materials Processing, HESAM University, Châlons-en-Champagne, France

Mohamed El Mansori, Arts et Metiers Institute of Technology, Mechanics Surfaces and Materials Processing, HESAM Université, Châlons-en-Champagne, France and Texas A&M Engineering Experiment Station, Institute for Manufacturing Systems, College Station, TX, United States

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Introduction

Natural fiber composites (NFC) are in increased demand from automotive and aerospace industries (Akampumuza *et al.*, 2017; Pandey *et al.*, 2010; Shalwan and Yousif, 2013). Natural fibers can compete with synthetic glass fibers in terms of mechanical (Dittenber and GangaRao, 2012; Pickering *et al.*, 2016), thermal and acoustic properties (Alves *et al.*, 2010; Etaati *et al.*, 2014; Rajeshkumar and Hariharan, 2014). Moreover, natural fibers are biodegradable and recyclable which make them eco-friendly and suitable for both circular economy and sustainable development (Ramesh *et al.*, 2017). NFC considered here are the assembly of natural fibers and polymeric matrices. If the polymer matrix is also bio-based, the NFC can be also called a green composite.

Industrial applications require high finishing quality of NFC parts, which is difficult to achieve with the usual elaboration processes such as the thermocompression technique for long continuous fiber composites (Davim and Reis, 2005; Nassar *et al.*, 2017). Indeed, NFC parts elaborated with thermocompression have usually a poor surface quality on the edges and should be reworked. Furthermore, some specific geometries such as holes for assembly cannot be gotten directly from the thermocompression. Therefore, machining operations are essential for this type of material in order to transform the NFC part to its final industrial application.

The microstructure of natural fibers is completely different from that of synthetic glass fibers (Baley, 2002; Morvan *et al.*, 2003). Consequently, the thermomechanical response of NFC is not similar to that of glass fiber composites (Khan *et al.*, 2011). The machining behavior of NFC requires hence a new investigation. For this aim, this article presents an exploratory study that investigates the machining behavior of NFC for different natural fibrous structures, different machining processes, and different machining parameters. This will provide to the reader a global idea about the machinability of these eco-friendly materials.

Natural Fiber Structure: From Plant Stem to Cellulose Microfibrils

The main natural fibers used in the composite industry are bast fibers extracted from the plant stem such as flax and hemp (Yu, 2015). The use of natural bast fibers is due to their high mechanical performances shown above all in their natural role of ensuring the rigidity of the plant stem.

Natural fibers are present in the plant stem in the form of a bundle of elementary fibers as shown in Fig. 1. These fiber bundles are extracted after retting of plant stem with a specific mechanical process of breaking and scutching (Sadrmanesh and Chen, 2019; Zimniewska *et al.*, 2011). Then, the fiber bundles are hackled to separate the elementary fibers as much as

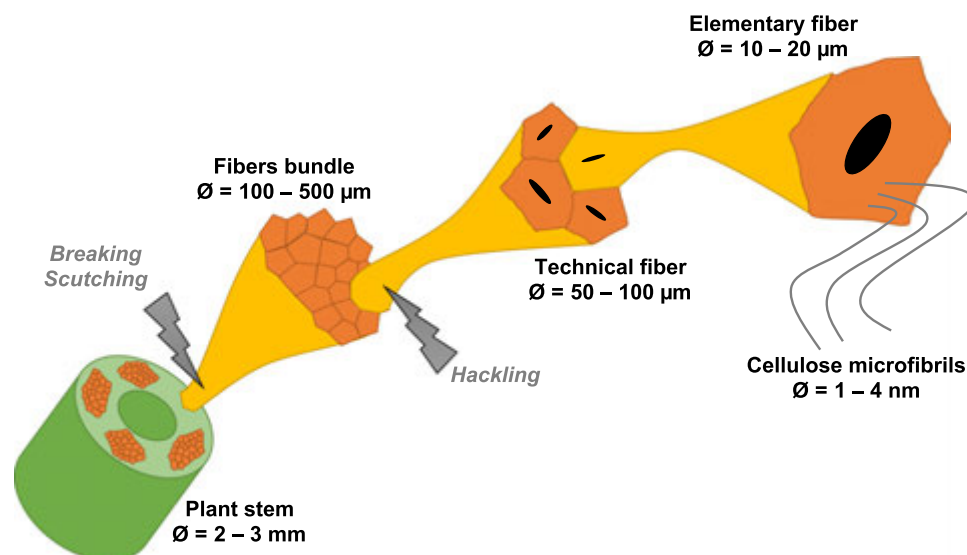


Fig. 1 Schematic illustration of bast fiber structure from their plant stem to cellulose.

possible. The resulting fibers are called technical fibers which are in form of reduced bundles of 3–6 elementary fibers. The elementary fiber has a cellulosic structure in form of a stacking of cellulosic cell walls (Baley, 2002). Each cell wall is itself a composite structure at nanoscale with cellulose microfibrils embedded in amorphous matrix mainly made of pectin and hemicellulose (Charlet *et al.*, 2007).

Experimental Methodology

Natural Fibrous Reinforcements

In this exploratory study, the main fibrous reinforcement structures commonly used in composites industry are investigated as shown in Fig. 2:

- Random distributed short fibers:
 - (1) Bamboo fiber composites (Bamboo FC).
 - (2) Miscanthus fiber composites (Miscanthus FC).
 - (3) Sisal fiber composites (Sisal FC).
- Unidirectional long fibers: Unidirectional flax fiber composites (UD flax FC).
- Bidirectional long fibers: Bidirectional flax fiber composites (BD flax FC).

Short fibers have an average length of 1 mm (Fig. 2(a)–(c)). The unidirectional flax reinforcement is performed with twisted fiber yarns. The unidirectionality of flax yarns is insured by weft synthetic fibers (Fig. 2(d)). For BD flax FC, the flax fiber reinforcement is in the form of 4×4 plain weave of the twisted flax yarns (Fig. 2(e)).

All the considered composite structures have a similar polymer matrix of polypropylene. Short fiber composites are elaborated using the thermo-injection process while the long fiber composites are manufactured with the them-compression technique. Table 1 presents the principal mechanical properties of the studied composites and their corresponding fibers.

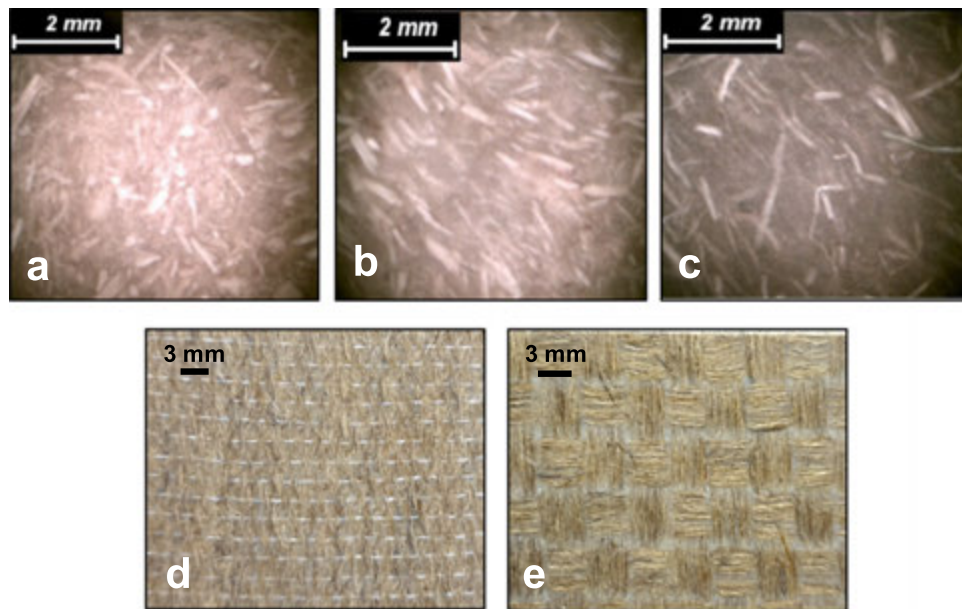


Fig. 2 Photographic images of each considered NFC. (a) Bamboo FC, (b) Miscanthus FC, (c) Sisal FC, (d) UD flax FC, and (e) BD flax FC.

Table 1 Mechanical properties of the considered NFC

	<i>PP bamboo</i>	<i>PP miscanthus</i>	<i>PP sisal</i>	<i>PP UD flax</i>	<i>PP BD flax</i>
<i>Elastic modulus (GPa)</i>	4.1	2.7	2.2	17.6	8.1
<i>Tensile strength (MPa)</i>	40	30	28	109	56
<i>Elastic modulus of fibers (GPa)</i>	19	13.8	7.84	50	50
<i>Tensile strength of fibers (MPa)</i>	89.2	62	50	500	500

Machining Processes

Fig. 3 summarizes the machining processes investigated in this study that are the traditional techniques commonly used in composite industry such as drilling and milling. The milling process is used to rework and resize the edge of the composite parts, while the drilling process is used to create the assembly holes on the composite parts. Moreover, the fundamental process of orthogonal cutting is also used to understand the elementary cutting mechanisms of this kind of materials.

Drilling and milling were performed using a CNC machine, while the orthogonal cutting was conducted with a shaper machine. Each machining process is instrumented with a piezoelectric dynamometer to apprehend the in-situ cutting forces. In addition, the orthogonal cutting device is equipped with a fast camera to capture the in-situ chip morphology.

Machining Analyses

Machined surfaces are analyzed in terms of microscopic surface state and surface topography. The microscopic surface state observations are performed with a scanning electron microscope (SEM) at low vacuum mode to investigate the cutting behavior of the natural elementary fibers within the composite structure. The resulting machined surfaces topography is quantified using a 2D stylus profilometer according to the ISO4287 standard, and 3D white light interferometer.

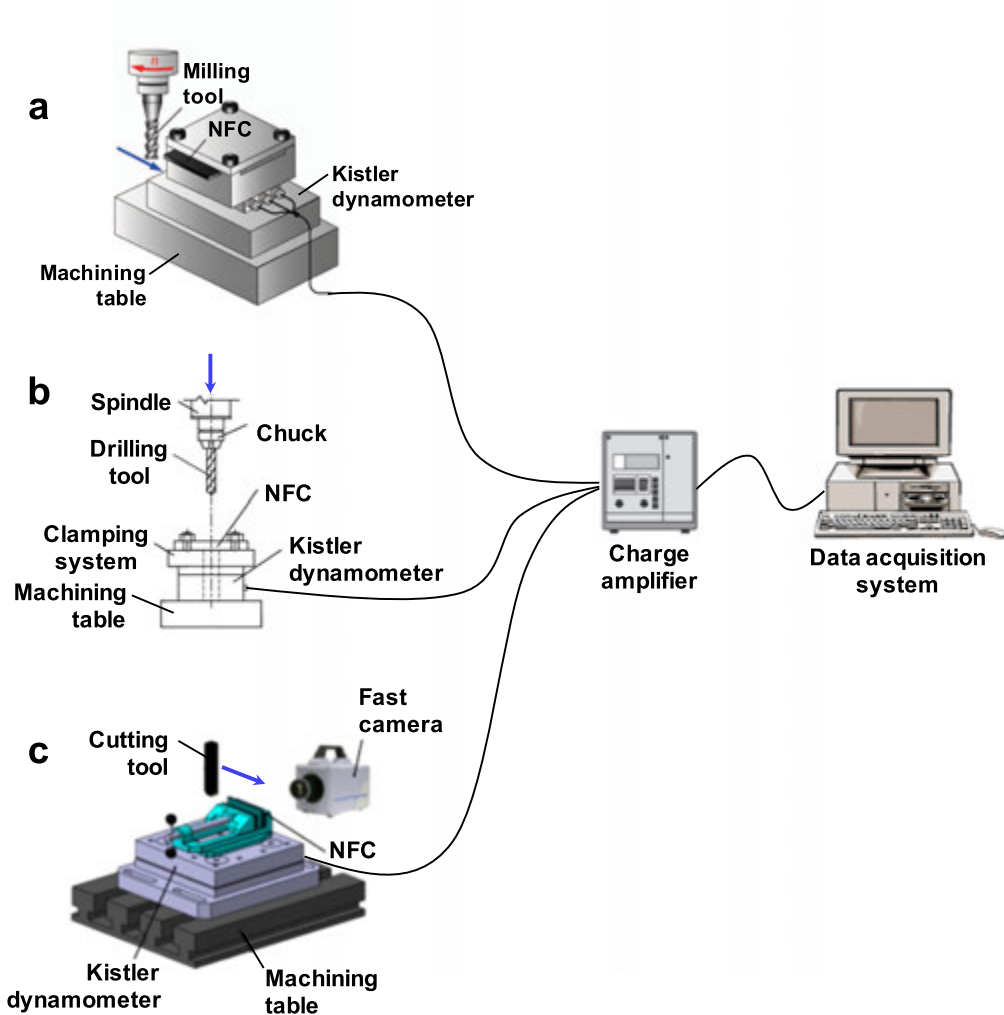


Fig. 3 Schematic illustration of the experimental machining setup. (a) milling, (b) drilling, and (c) orthogonal cutting.

Analysis of Machining Behavior of Natural Fiber Composites

Effect of Fiber Type

The effect of fiber type is investigated on the NFC reinforced with short fibers of bamboo, miscanthus and sisal using the milling process (Chegdani *et al.*, 2015b). All the machining parameters are kept constant except the feed that varies from 0.04 to 0.12 mm/tooth. The machining analysis is performed by:

- Calculating the specific cutting energy;
- Observing the machined surfaces by SEM;
- Measuring the machined surfaces topography using the 2D profilometer before and after the machining operation.

The effect of the advance will be discussed later in this article (Section “Effect of cutting feed”).

Fig. 4 presents the microscopic machined surface state of each NFC with the corresponding topographic signal at the same cutting conditions. Bamboo fiber composites show the best fiber shearing without significant breaking of interfaces (Fig. 4(a)). Miscanthus fiber composites have also a good fiber shearing but interfaces break is noticed, especially between the elementary fibers (Fig. 4(b)). For sisal fiber composites, it can be seen a significant deformation of fibers toward the feed direction before shearing. This induced uncut fiber extremities that remain on the machining surfaces in addition to important decohesion zones due to interfaces break (Fig. 4(c)).

Fig. 4 shows that the corresponding topographic signal of the machined surfaces reflects the phenomena observed in the SEM images. The topographic signal of Bamboo fiber composites shows the lowest irregularities (Fig. 4(d)) while that of sisal fiber composites reveals the highest irregularities (Fig. 4(f)). This indicates that the shearing of natural fibers differs in function of the fiber type and can be confirmed by analyzing the specific cutting energy of the three NFC as shown in Fig. 5(a). Indeed, the specific cutting energy of sisal fiber composites reveals the highest values while that of bamboo fiber composites shows the lowest. This means that the fiber shearing is the most effective in the case of bamboo and the least effective in the case of sisal. Consequently, cutting NFC with sisal fibers induces high surface roughness as shown in Fig. 5(b) because of uncut fibers extremities and interfaces break. This cutting behavior of natural fibers is due to their mechanical properties, especially the fiber rigidity that controls the cutting contact stiffness. As shown in Table 1, bamboo fibers have the highest rigidity and sisal fibers express the lowest one.

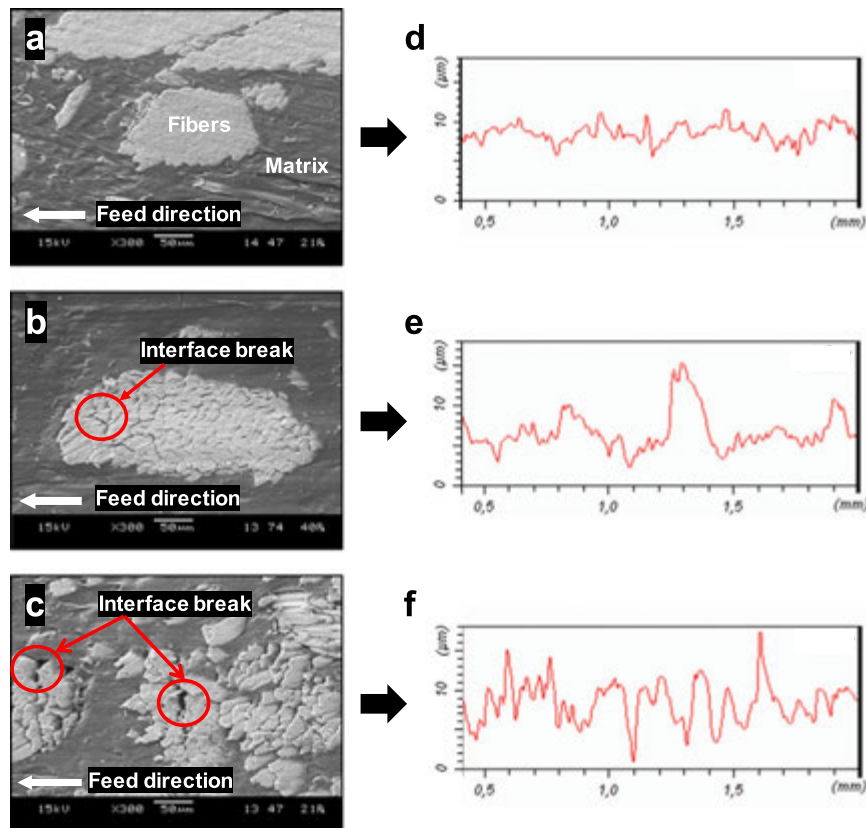


Fig. 4 Typical SEM images and their corresponding topographic signals of machined surfaces for (a, d) Bamboo FC, (b, e) Miscanthus FC, and (c, f) Sisal FC.

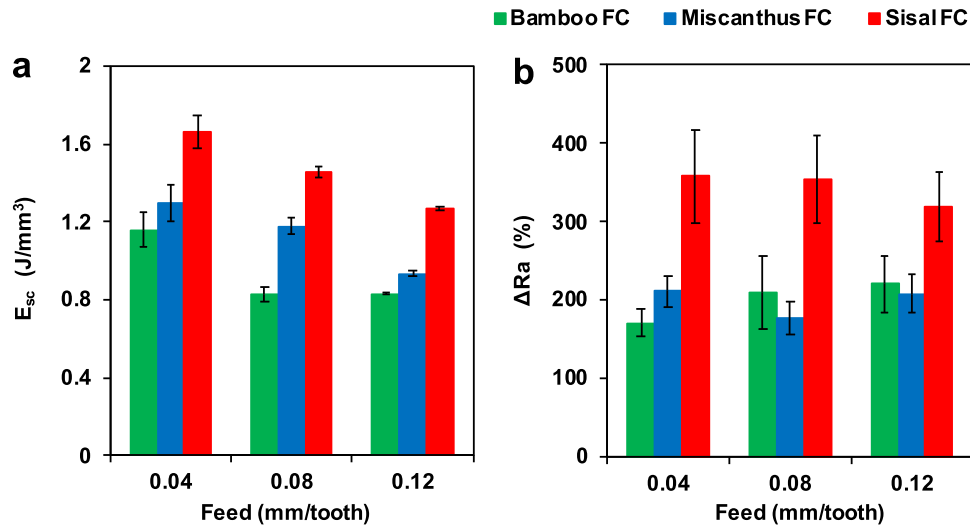


Fig. 5 (a) Specific cutting energy of the short NFC. (b) Roughness gain ratio of the machined surfaces for the short NFC.

Therefore, for next sections, the investigations will be performed on flax fiber reinforced polypropylene composites since flax fibers have the highest elastic modulus comparing to the other natural fiber types.

Effect of Fiber Orientation

In this section, UF flax FC are considered. The orthogonal cutting process is used in order to isolate the fiber orientation effect. The three principal fiber orientation with respect to the cutting direction are investigated ($\theta = 0^\circ$, $\theta = 45^\circ$, and $\theta = 90^\circ$). Only the effect of fiber orientation is discussed in this section.

Fig. 6 shows that the cutting behavior of flax fibers differs by changing their orientation relative to the cutting direction. Cutting with $\theta = 90^\circ$ induces significant uncut fiber extremities that remain on the machined surface (**Fig. 6(a)**). The fiber shearing is the most effective with $\theta = 45^\circ$ where the uncut fiber extremities are not obvious on the machined surface (**Fig. 6(b)**). For $\theta = 0^\circ$, the cutting behavior is random where flax fibers are either sheared, detached or torn-off (**Fig. 6(c)**) which is related to the random cutting contact location. This random cutting behavior induces the highest surface roughness as shown in **Fig. 7(b)**. The non-effective fiber shearing in the case of $\theta = 90^\circ$ is noticed with the highest shearing energy as shown in **Fig. 7(b)** while the cutting configuration with $\theta = 45^\circ$ induces the lowest cutting energy and the lowest machined surface roughness. The poor fiber shearing when $\theta = 90^\circ$ is due to the high transverse elasticity of natural fibers that favors the transverse deformation during the contact with the cutting tool. For $\theta = 45^\circ$, the shearing effectiveness is related to the fact that the fiber orientation (i.e., the highest stiffness direction) is toward the shearing plan of the cutting system.

Effect of Tool Geometry

Effect of cutting edge radius

In this section, milling and drilling processes are considered. For both processes, the variation of the cutting edge radius is controlled by the tool coating. Therefore, the lowest cutting edge radius is obtained with the uncoated cutting tool. Then, titanium diboride (TiB_2) and diamond coatings are applied to increase the cutting edge radius. TiB_2 coating is performed by monolayer physical vapor deposition (PVD) while diamond coating is achieved using multilayer chemical vapor deposition (CVD). Consequently, the two coating depositions techniques generate different coating thicknesses and lead to vary the cutting edge radius. **Table 2** summarizes the tools coatings characteristics.

• Effect of cutting edge radius in milling process

The investigation of cutting edge radius effect in milling process is performed on UD flax FC (**Chegdani et al., 2015a**). The SEM images of **Fig. 8** reveal the high impact of the cutting edge radius on the machining behavior of NFC. Flax fibers are well sheared with the uncoated milling tool that has the sharpest cutting edge (**Fig. 8(a)**). Increasing the cutting edge radius by about $6 \mu m$ changes completely the cutting behavior of flax fibers where a high rate of uncut fiber extremities are noticed in the machined surfaces (**Fig. 8(c)**). This shows that the shearing effectiveness of fibers is significantly affected, and the cutting behavior of flax fibers is highly sensitive to the small variation of the cutting edge radius.

The effect of the cutting edge radius on the shearing behavior of UD flax FC is also depicted in the energetical analysis of **Fig. 9(a)** where the highest edge radius induces the highest specific cutting energy. Consequently, the uncut fiber extremities caused by the poor fiber shearing generates the highest surface roughness as shown in **Fig. 9(b)**.

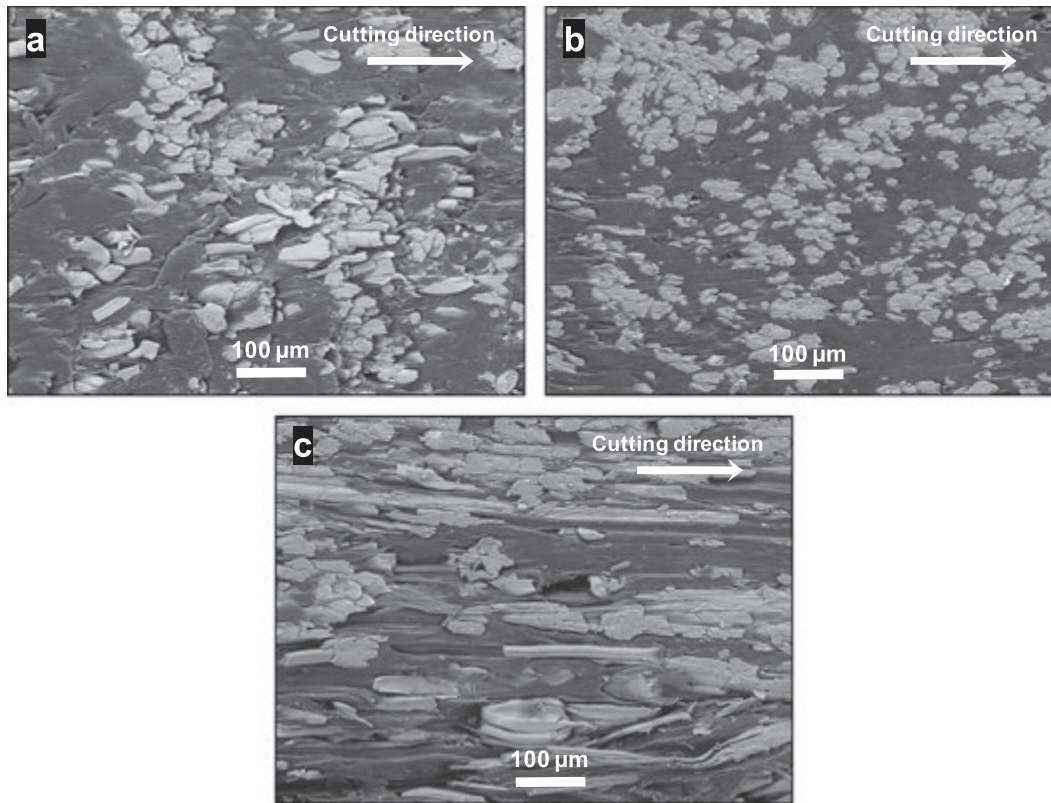


Fig. 6 SEM images of the machined surfaces of UF flax FC with different fiber orientation. (a) $\theta = 90^\circ$, (b) $\theta = 45^\circ$, and (c) $\theta = 0^\circ$.

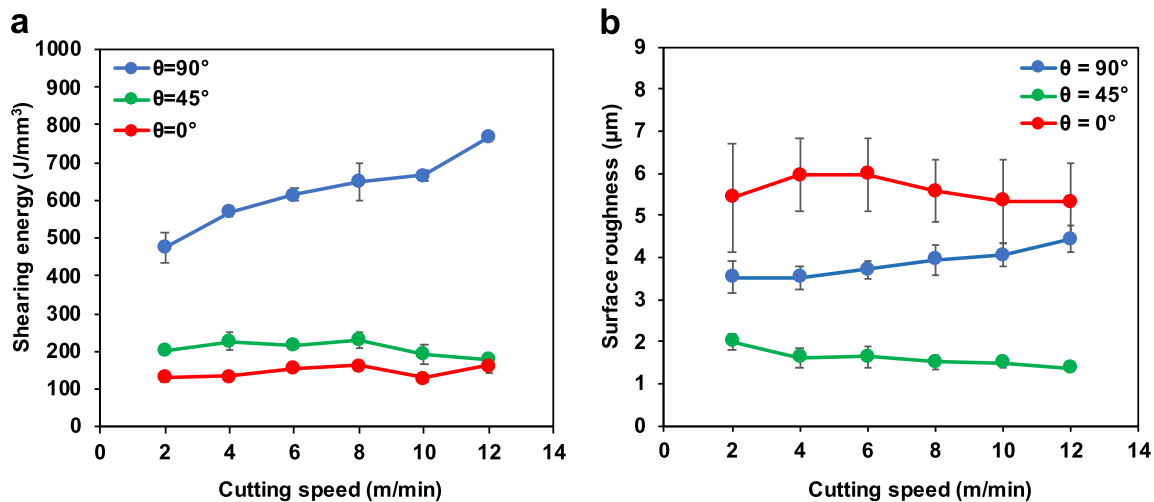


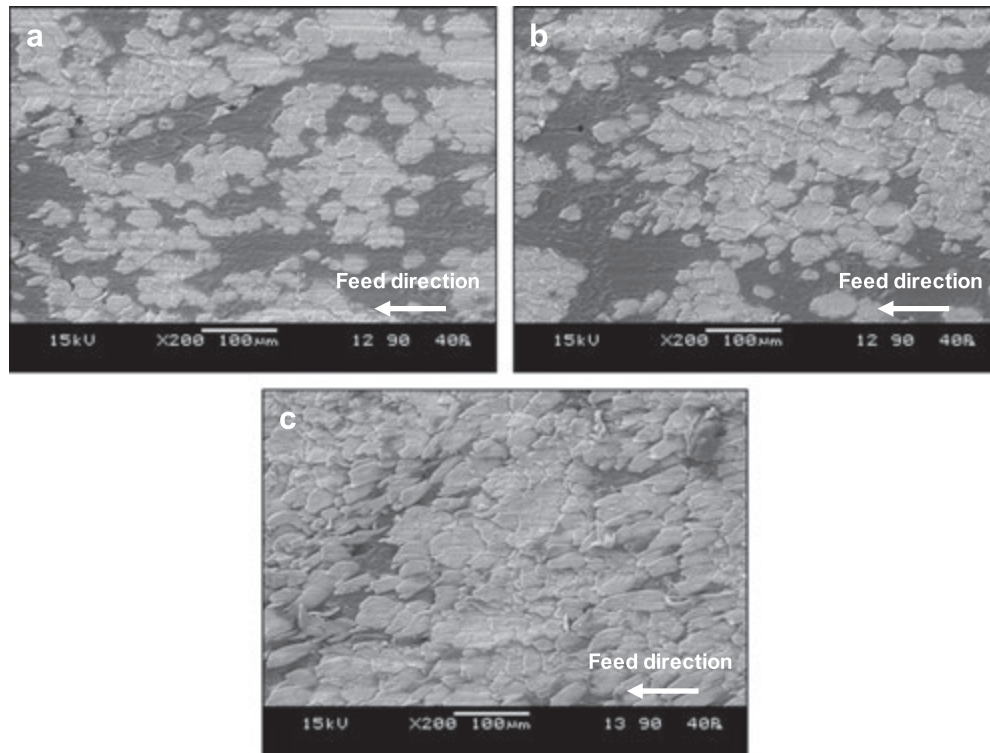
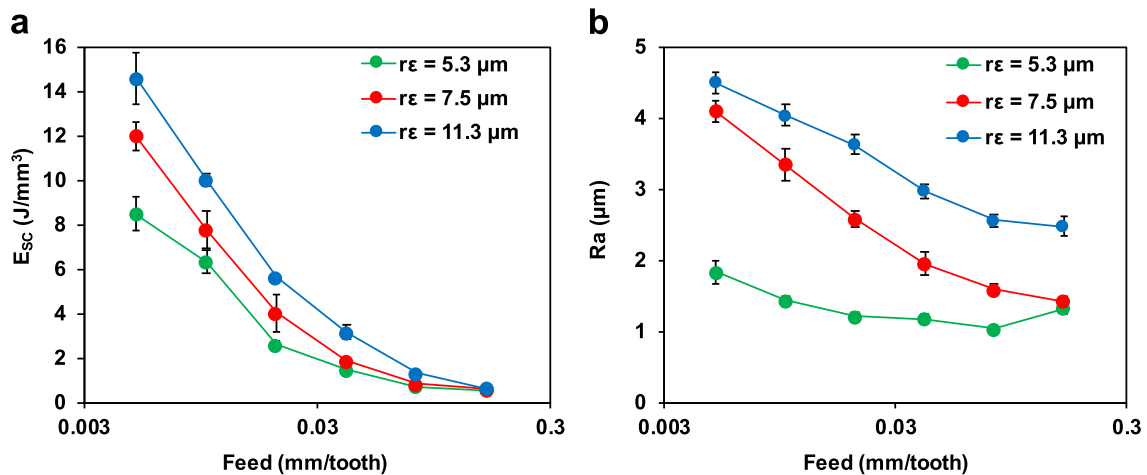
Fig. 7 (a) Specific shearing energy of UD flax FC for different fiber orientations. (b) Arithmetic surface roughness of machined surfaces of UD flax FC for different fiber orientations.

• *Effect of cutting edge radius in drilling process*

For the drilling process, the analysis of the cutting edge radius effect is performed on BD flax FC (Chegdani and El Mansori, 2018). The cutting behavior of flax fibers during the drilling operation shown in Fig. 10 is similar to that of the milling process presented previously in Fig. 8. Indeed, drilling with the lowest cutting edge radius enhance the shearing behavior of flax fibers as shown in Fig. 10(a) where the fibers cross sections are well perceptible. Flax fibers start to deform transversely when increasing the cutting edge radius from 5.5 μm to 8 μm . Uncut fiber extremities are then noticeable in the machined surface of Fig. 10(b). When drilling with a cutting edge radius of 11.5 μm , the cutting behavior is completely changed from shearing to torn-off as clearly shown in Fig. 10(c).

Table 2 Coating characteristics of drilling and milling tools

		Uncoated	TiB ₂ coated	Dia. coated
Milling tools	Substrate	carbide	carbide	carbide
	Coating thickness (μm)	–	2 ± 0.7	7 ± 1
	Measured cutting edge radius (μm)	5.3 ± 0.6	7.5 ± 0.5	11.3 ± 0.7
Drilling tools	Substrate	carbide	carbide	carbide
	Coating thickness (μm)	–	2 ± 0.7	7 ± 1
	Measured cutting edge radius (μm)	5.5 ± 0.5	8 ± 0.5	11.5 ± 1

**Fig. 8** SEM images of the milled surfaces of UD flax FC with different values of cutting edge radii. (a) $r_\epsilon = 5.3 \mu\text{m}$, (b) $r_\epsilon = 7.5 \mu\text{m}$, and (c) $r_\epsilon = 11.3 \mu\text{m}$.**Fig. 9** (a) specific cutting energy, and (b) Arithmetic surface roughness of milled surfaces for UD flax FC with different values of cutting edge radii.

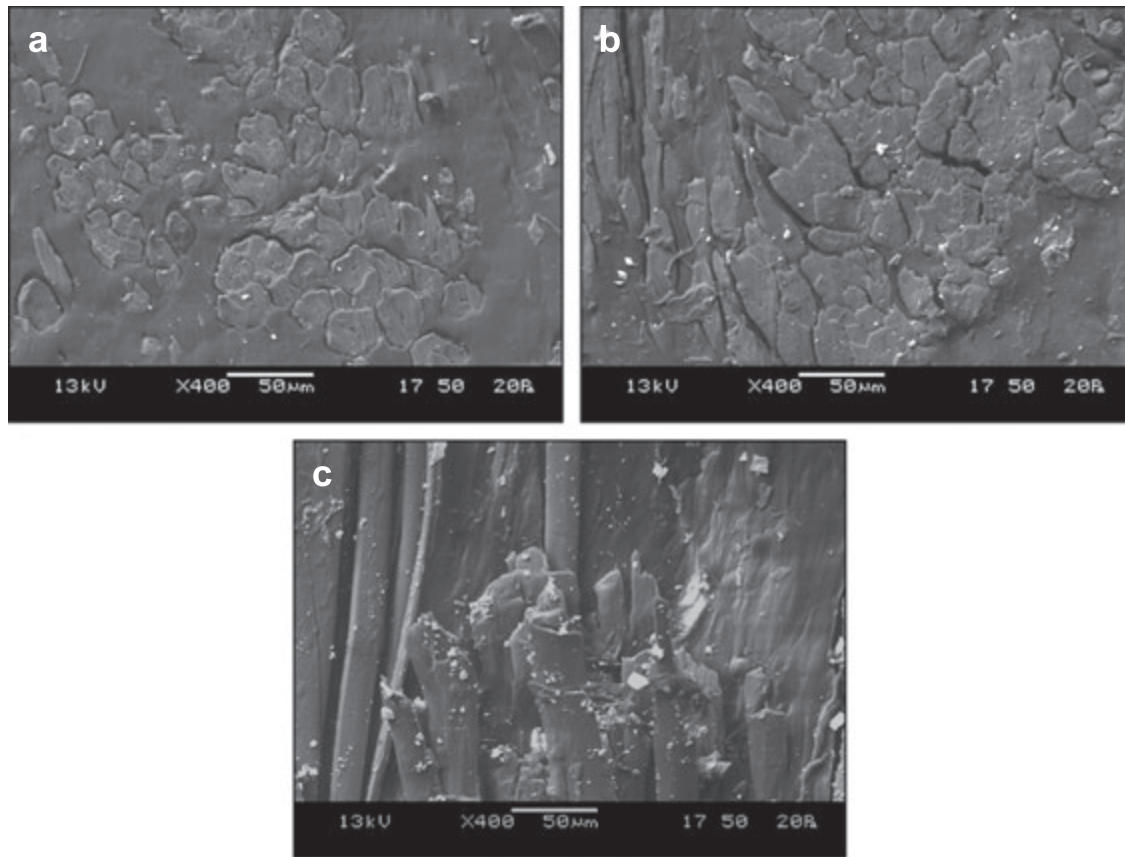


Fig. 10 SEM images of the drilled surfaces of BD flax FC with different values of cutting edge radii. (a) $r_\epsilon = 5.5 \mu\text{m}$, (b) $r_\epsilon = 8 \mu\text{m}$, and (c) $r_\epsilon = 11.5 \mu\text{m}$.

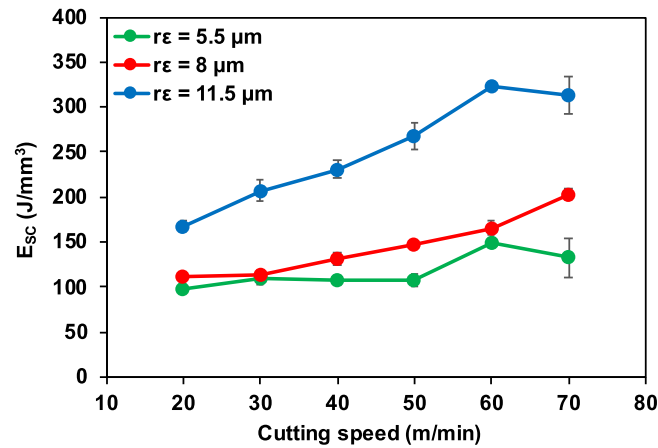


Fig. 11 Specific cutting energy of drilled surfaces for BD flax FC with different values of cutting edge radii.

The cutting behavior of flax fibers inside the composite is also exposed in the measured cutting energy as shown in Fig. 11. The specific cutting energy of the drilling operation with a cutting edge radius of $11.5 \mu\text{m}$ is largely higher than the other configurations. This shows once again how small variations of the cutting edge radius can affect the cutting behavior of NFC. This is due to the cutting contact scale, specifically the contact scale between the cutting edge and the natural fibers. The diameter of elementary flax fibers is between $10 \mu\text{m}$ and $20 \mu\text{m}$, which is in the same order of magnitude than the cutting edge radius. Therefore, when the cutting edge radius value approaches the diameter of the fiber, this latter is more likely to deform than to shear. If the cutting edge radius is greater than or equal to the fiber diameter, the fiber torn-off become the main cutting mechanism.

Effect of helix angle

The effect of the helix angle is investigated through a milling process of BD flax FC (Chegdani *et al.*, 2016). Three milling tools with similar geometrical parameters, except the helix angle of the cutting edges, have been considered. The helix angles values vary from $H = 0^\circ$ to $H = 40^\circ$. Since the cutting edge radius increase impacts negatively the cutting behavior of fibers (Section “Effect of cutting edge radius”), uncoated milling tool are chosen in this study.

The SEM images of Fig. 12 discriminate two fiber zones. Warp fiber zones (WPZ) is the zone with flax fibers that are oriented perpendicularly to the feed direction, while weft fiber zone (WTZ) is the zone with flax fibers that are oriented toward the feed direction. As for the fiber orientation of $\theta = 0^\circ$ in Section “Effect of Fiber Orientation”, the cutting behavior of flax fiber in the weft fiber zone is random and depends on the cutting contact location between the cutting edge end and the fibers. However, the warp fiber zone shows an effect of the helix angle on the cutting behavior of flax fibers. Cutting with zero helix angle offers the best shearing of flax fibers as shown in Fig. 12(a) where the cross section of fibers is visible on the microscopic images of the machined surfaces. The shearing effectiveness decreases when increasing the helix angle as shown in Fig. 12(b) and (c) because flax fibers on the warp fiber zone are deformed toward the feed direction and uncut fiber extremities are noticeable on the machined surfaces. This cutting behavior of flax fibers in function of the tool helix angle can be explained by the behavior of the cutting forces presented in Fig. 13. Indeed, cutting with zero helix angle concentrates the machining forces in the feed direction which provides a high contact stiffness for cutting (Fig. 13(a)). When increasing the helix angle, the machining forces are reoriented from the feed direction to the tool axis direction until becoming similar for $H = 40^\circ$. This weakens the shearing of flax fibers and favors their deformation.

The effect of tool helix angle on the cutting behavior of flax fibers is revealed in the topographic analysis of the machined surfaces of BD flax FC by quantifying the arithmetic surface roughness on the warp fiber zones and the weft fiber zones separately as shown in Fig. 14. On the warp fiber zone, increasing the tool helix angle increases the surface roughness because of the uncut fiber extremities (Fig. 14(a)). However, the random cutting behavior of flax fibers on the weft fiber zones induces a random trend of the surface roughness evolution with high variability in the measurements (Fig. 14(b)).

Effect of rake angle

The effect of rake angle is investigated on UD flax FC using the orthogonal cutting process (Chegdani *et al.*, 2018b). All the cutting parameters are kept constant, except the rake angle (γ) of the cutting toll that varies from -4.5° to 5.5° .

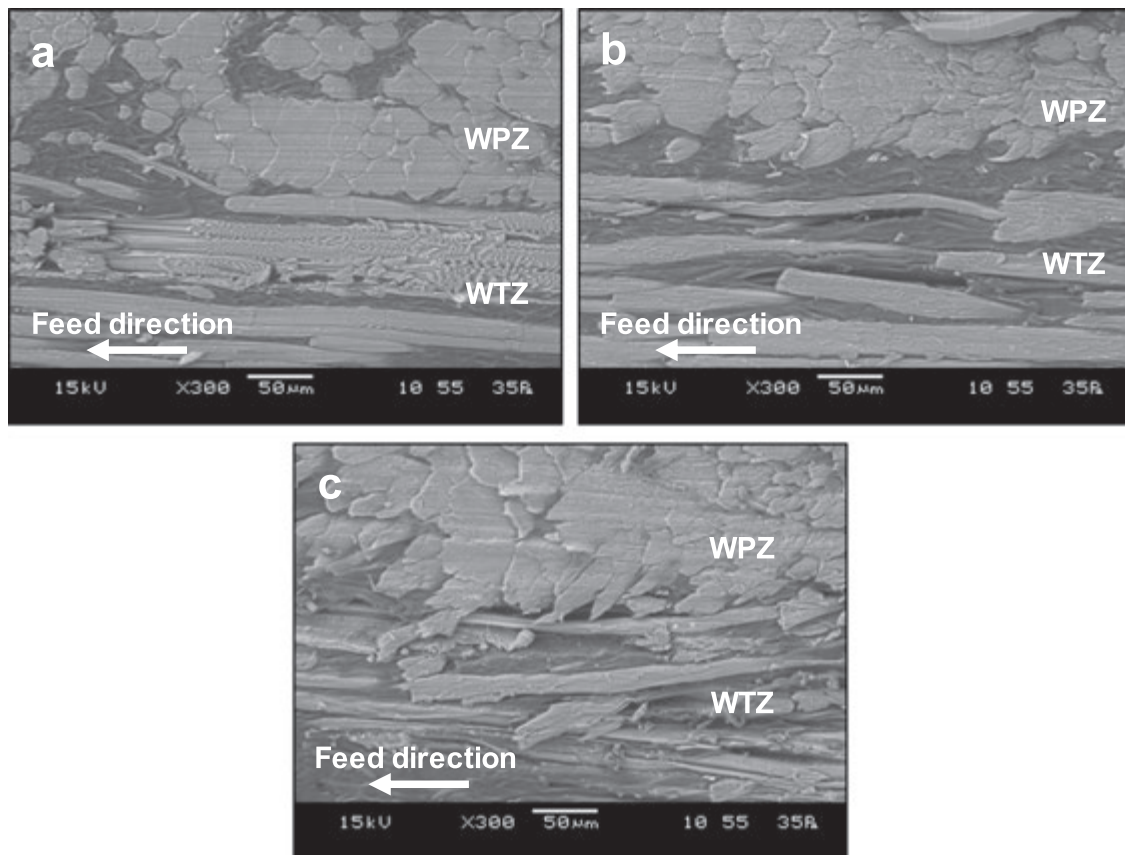


Fig. 12 SEM images of the milled surfaces of BD flax FC with different values of helix angles. (a) $H = 0^\circ$, (b) $H = 20^\circ$, and (c) $H = 40^\circ$.

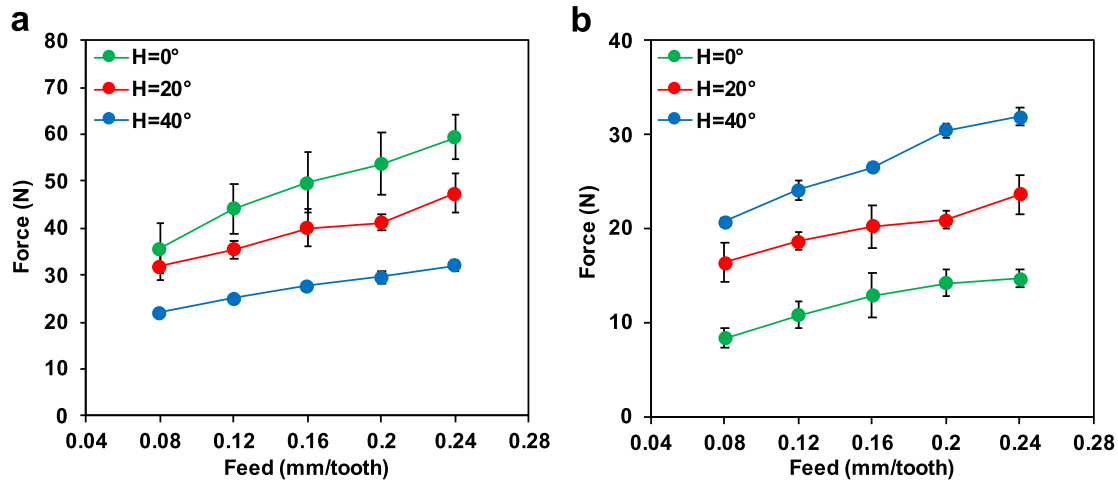


Fig. 13 Machining forces of BD flax FC with different values of helix angles. (a) forces in the feed direction, and (b) forces in the tool axis direction.

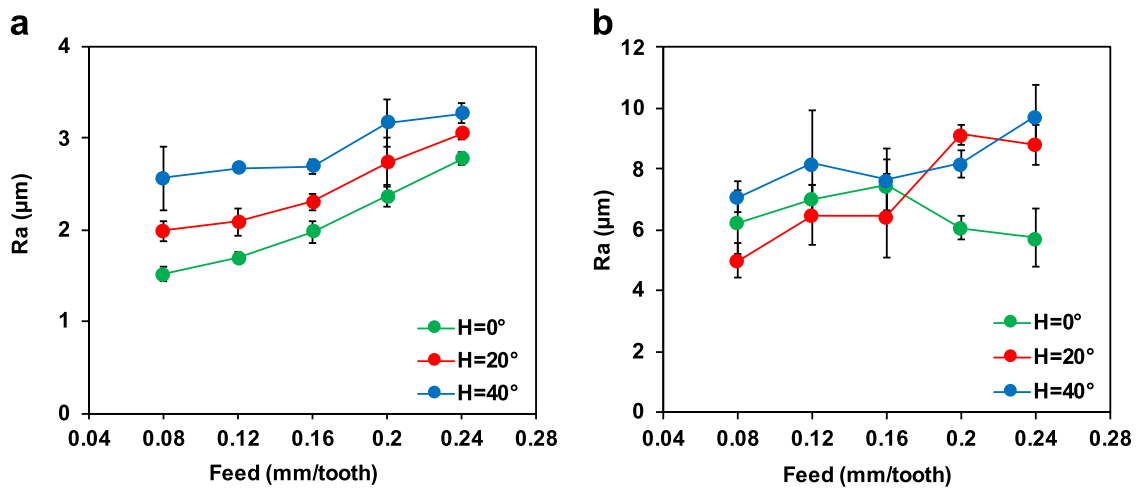


Fig. 14 Arithmetic mean of the surface roughness of the machined surfaces of BD flax FC with different values of helix angles. (a) on warp fiber zones, and (b) on weft fiber zones.

The SEM images of Fig. 15 does not show a significant difference on the microscopic surface state of the machined surfaces performed with the different values of rake angle. However, a slight enhancement of the fiber shearing is noticed on the machined surfaces achieved with positive rake angles (Fig. 15(c) and (d)). Concretely, negative rake angles cause transverse deformation of flax fibers toward the cutting direction before shearing.

On the other hand, changing the tool rake angle affects the chip morphology in terms of chip curling. As shown in Fig. 16, cutting with negative rake angle avoid the curling of the chip during its formation (Fig. 16(a)). Increasing the rake angle from negative to positive values favors the chip curling (Fig. 16(c)). Even if the fast-cam images of Fig. 16 were taken at the same cutting time, it can be seen that the chip length of negative rake angle is the smallest and increases with rake angle increase. Indeed, negative rake angle compress the material toward the shear zone which cause a plastic deformation of the removed chip by compression. Consequently, the chip thickness increases, and the chip length decreases.

The tool rake angle affects significantly the cutting force. As shown in Fig. 17(a), increasing the rake angle from -4.5° to 5.5° generates a 76% increase in cutting force. This reveals the importance of considering the rake angle effect for the design of the machining operations of NFC in order to prevent the tool wear.

The effect of rake angle presented on the SEM images of Fig. 15 is reproduced with the topographic analysis of the machined surfaces as shown in Fig. 17(b). Indeed, the mean arithmetic surface roughness of the machined surfaces decreases slightly by increasing the rake angle from -4.5° to 5.5° .

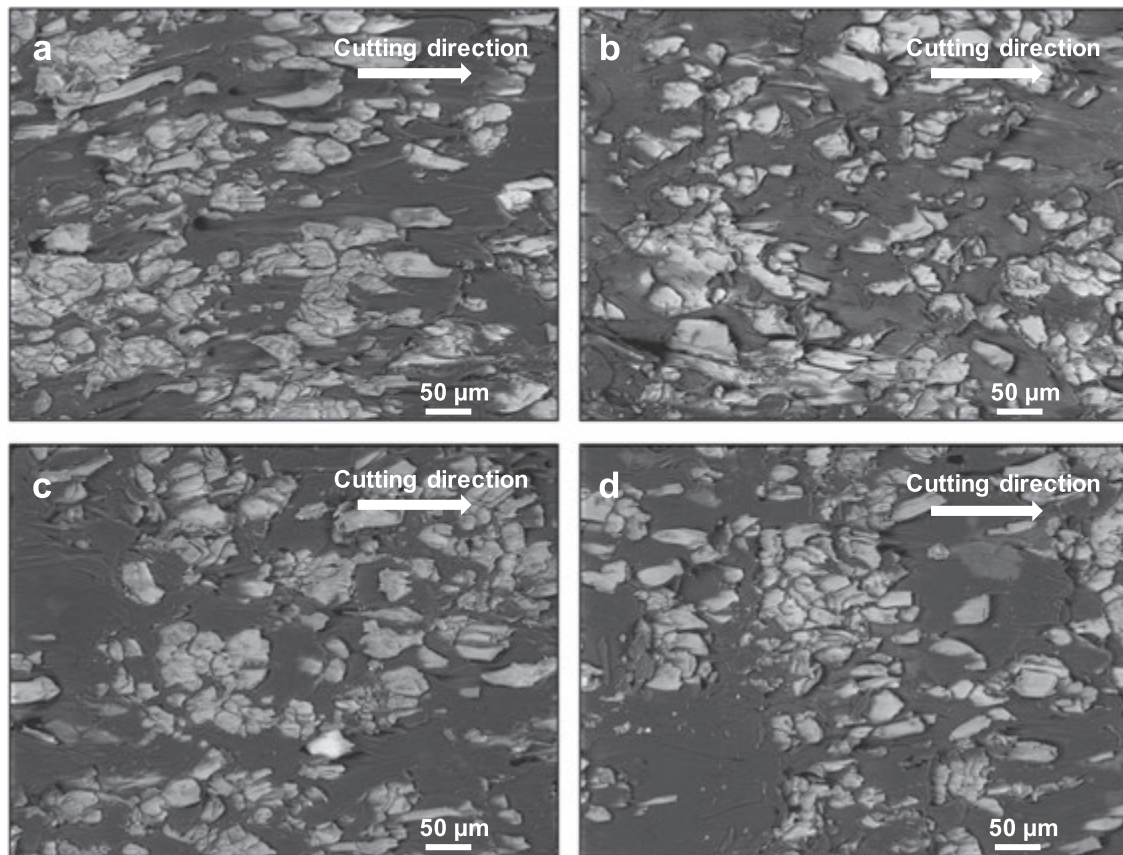


Fig. 15 SEM images of the machined surfaces of UD flax FC with different values of rake angles. (a) $\gamma = -4.5^\circ$, (b) $\gamma = 0^\circ$, (c) $\gamma = 3^\circ$, and (d) $\gamma = 5.5^\circ$.

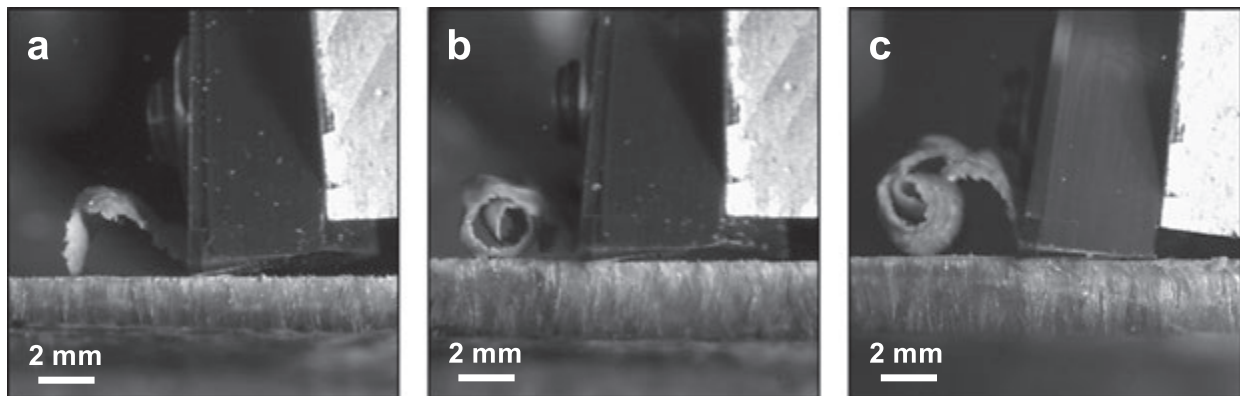


Fig. 16 Fast-cam images on the same cutting time of the chip formation generated with different values of rake angles. (a) $\gamma = -4.5^\circ$, (b) $\gamma = 0^\circ$, and (d) $\gamma = 5.5^\circ$.

Effect of Process Parameters

Effect of cutting speed

The effect of cutting speed was presented previously in [Fig. 7](#) for the orthogonal cutting process of UD flax FC. Small cutting speed values were used in this investigation to avoid heat generation. [Fig. 7\(a\)](#) shows that the cutting speed impacts the machining of NFC with fibers oriented perpendicularly to the cutting direction. In this cutting configuration, increasing the cutting speed increases the shearing energy of the cutting process. However, the effect of the cutting speed the surface topography is not significant ([Fig. 7\(b\)](#)). The same observation is noticed in the case of the drilling process as shown in [Fig. 11](#) where increasing the cutting speed increases the cutting energy.

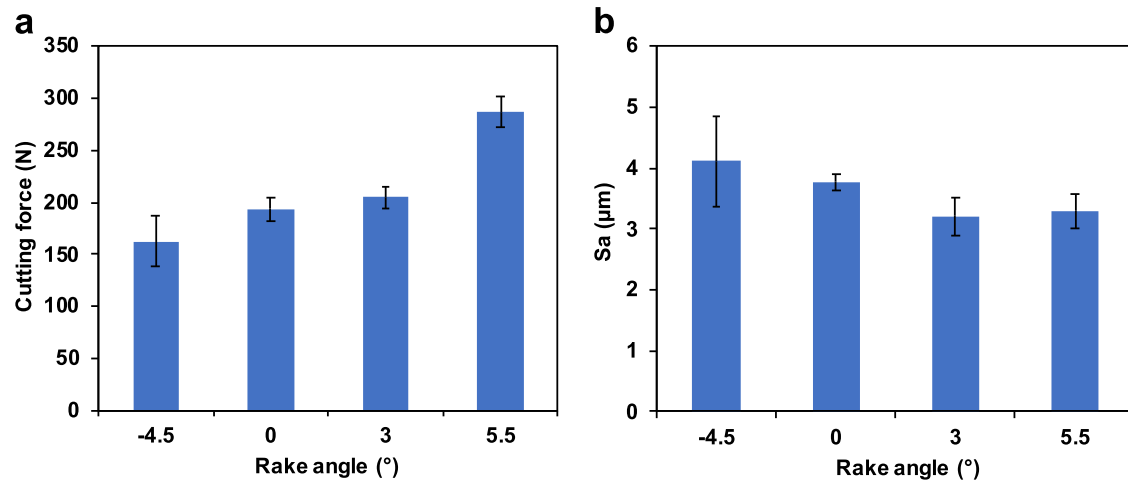


Fig. 17 (a) Cutting force of UD flax FC with different values of tool rake angles. (b) 3D arithmetic mean surface roughness of the machined surfaces of UD flax FC with different values of tool rake angles.

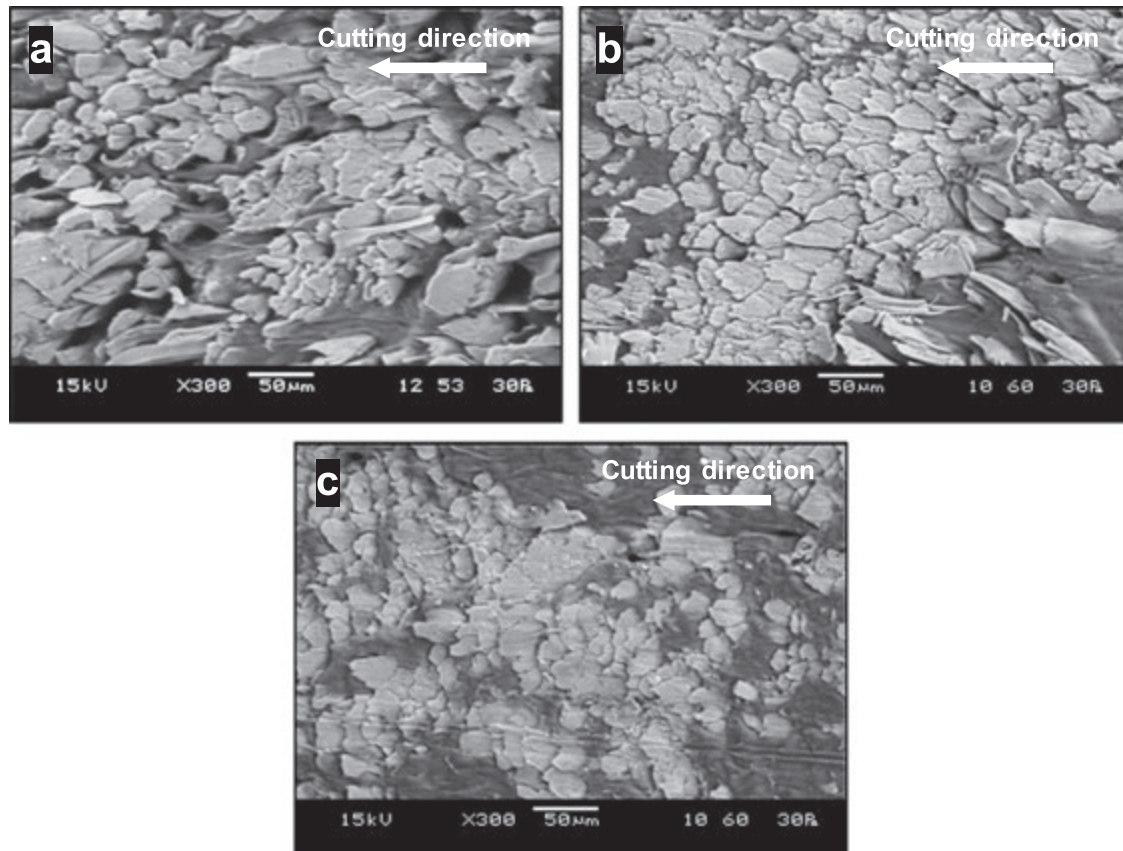


Fig. 18 SEM images of the machined surfaces of UD flax FC with different values of cutting speed. (a) $V_c = 12$ m/min, (b) $V_c = 32$ m/min, and (c) $V_c = 50$ m/min.

The effect of cutting speed is investigated in depth using orthogonal cutting of UD flax FC and realistic cutting speed values from 12 m/min to 80 m/min (Chegdani and El Mansori, 2018a). The SEM images of Fig. 18 show the important role of the cutting speed for the enhancement of the shearing effectiveness of flax fibers. Indeed, increasing the cutting speed from 12 m/min to 80 m/min reduces significantly the uncut fiber extremities and the decohesion zones caused by the damage of the interfaces.

Regarding the machining forces, it can be seen from Fig. 19 that the effect of cutting speed is similar to that observed in Fig. 7 and Fig. 11 where increasing the cutting speed increases the machining forces. The increase of the machining forces contributes

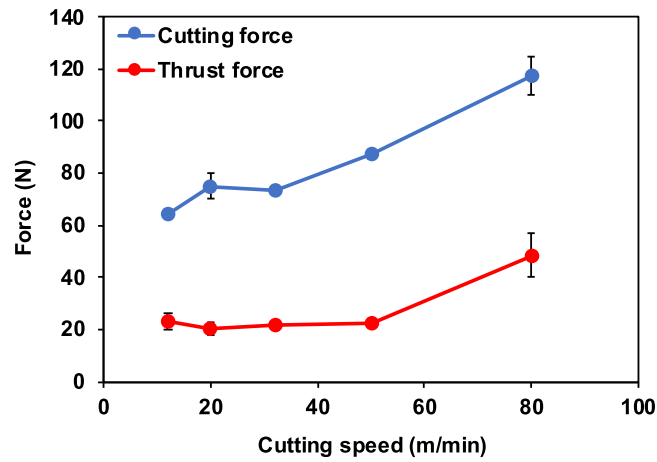


Fig. 19 Machining forces of the orthogonal cutting process of UD flax FC with different values of cutting speed.

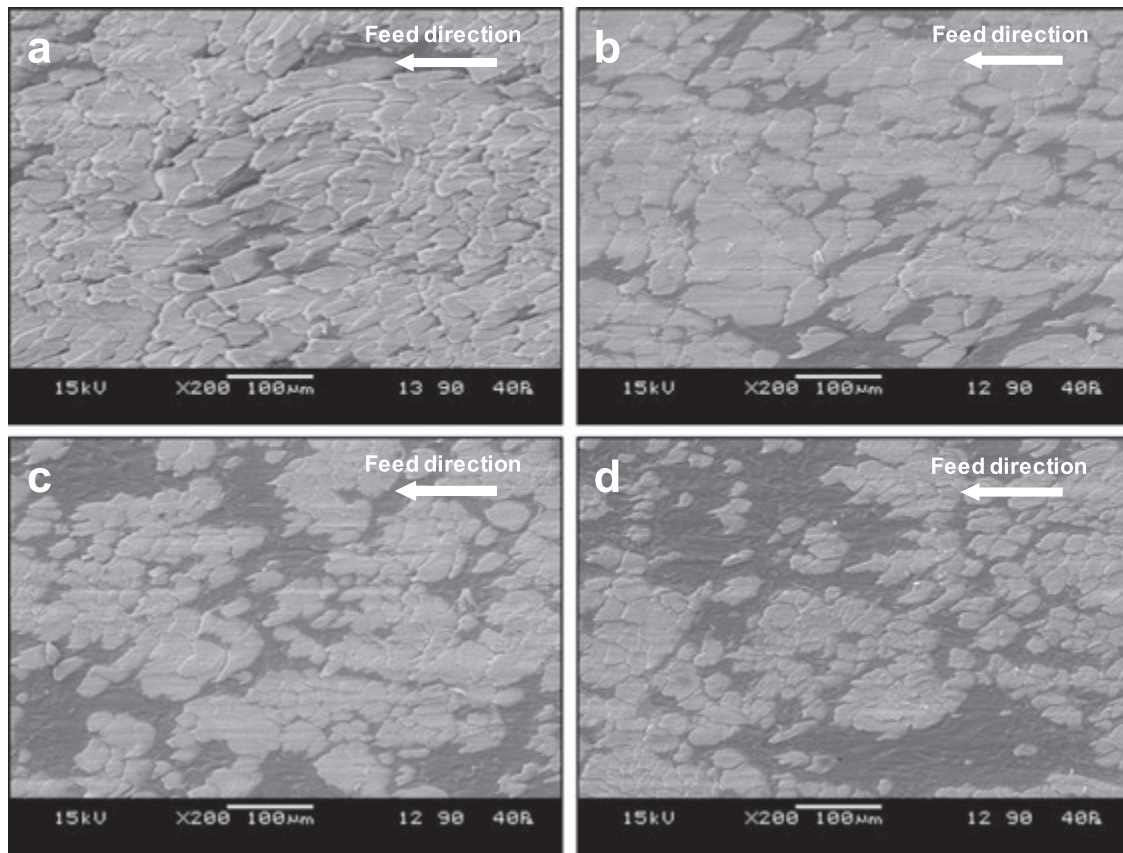


Fig. 20 SEM images of milled surfaces of UD flax FC performed with different values of cutting feed. (a) $f_z = 0.005$ mm/tooth, (b) $f_z = 0.02$ mm/tooth, (c) $f_z = 0.04$ mm/tooth, and (d) $f_z = 0.08$ mm/tooth.

then to the enhancement of the fiber shearing and the reduction of interface damages as shown in Fig. 18. It is important to notice that the effect of cutting speed on the machined surface state is activated at high speed values as shown in Fig. 7(b) where the effect of cutting speed is insignificant for speed values range from 2 m/min to 12 m/min.

Effect of cutting feed

The effect of cutting feed has been investigated for short fiber composites and long fiber composites using the milling process. In the case of short fiber composites (Fig. 5), increasing the feed from 0.04 mm/tooth to 0.12 mm/tooth decreases the specific cutting energy which leads to a decrease of the surface roughness of the machined surfaces.

In the study of the milling process of UD flax FC (Chegdani *et al.*, 2015a), the effect of the cutting feed has been investigated deeply by exploring a large range of feed values from 0.005 mm/tooth to 0.16 mm/tooth as shown in Fig. 9. The specific cutting energy of the milling process shows a significant decrease when increasing the cutting feed (Fig. 9(a)). This feed increase contributes also the decrease of the machined surface roughness (Fig. 9(b)).

For more understanding of the cutting feed effect, Fig. 20 shows the cutting behavior of flax fibers during the milling process of UD flax FC in function of the cutting feed. It is evident that increasing the feed enhance the fiber shearing and reduces significantly the uncut fiber extremities that remain on the machined surface. As well known in the machining fundamentals, the chip thickness is proportional to the cutting feed in the case of milling process. The chip thickness has to reach a minimum critical value to be in the favorable cutting conditions. Below this critical value, the cutting system is in the unfavorable cutting conditions that favor sliding and plastic deformation.

Effect of cutting depth

The effect of the cutting depth is investigated in the same study of the cutting speed presented in Section “Effect of cutting speed” where orthogonal cutting process is applied to UD flax FC (Chegdani and El Mansori, 2018a). The cutting depth is varied from 100 μm to 500 μm .

The fast-cam images of Fig. 21 show that the chip is curled and remains continuous all over the studied range of cutting depth. This finding was also noticed all over the studied range of cutting speed of the same investigation. Moreover, the removed chip remains also continuous when varying the fiber orientation as shown in the fac-cam images of Fig. 16. The continuity of the removed chip, regardless of the machining parameters, is principally due to the ductile behavior of both flax fibers and polypropylene matrix. The high transverse elasticity of flax fibers gives them the ability to deform easily and flow the deformation motion of the cutting without brittle fractures.

SEM observation of Fig. 22 reveal that increasing the cutting depth deteriorates the microscopic state of the machined surfaces. At low cutting depth ($a_p = 100 \mu\text{m}$), flax fibers show the best shearing of flax fibers with insignificant interface damages (Fig. 22(a)). However, the increase of the cutting depth from 100 μm to 500 μm intensifies the transverse deformation of flax fibers in addition to decohesion zones caused by the broken interfaces (Fig. 22(c)). This cutting behavior is depicted in the machining forces presented in Fig. 23 where a drastic increase of the cutting forces is noticed when varying the cutting depth from 100 μm to 500 μm . The thrust forces show a slight increase in function of the cutting depth.

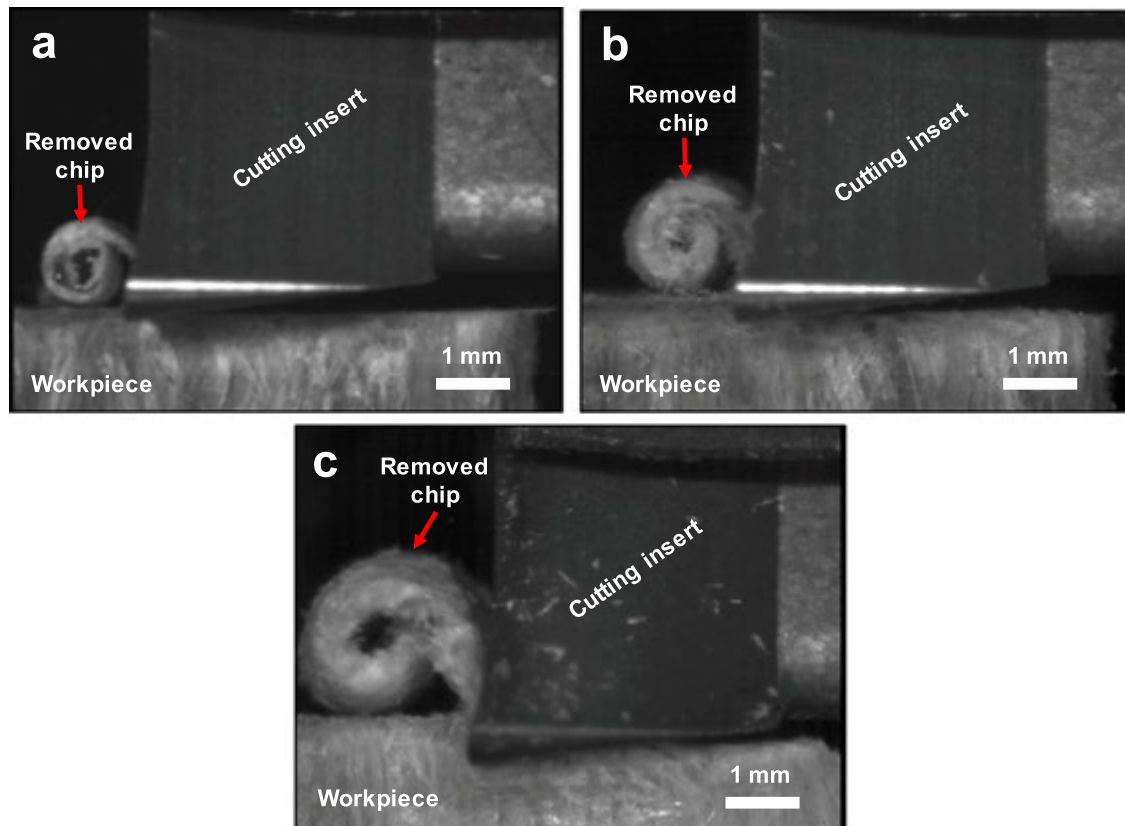


Fig. 21 Fast-cam images of the chip formation generated with different values of cutting depth. (a) $a_p = 100 \mu\text{m}$, (b) $a_p = 300 \mu\text{m}$, and (c) $a_p = 500 \mu\text{m}$.

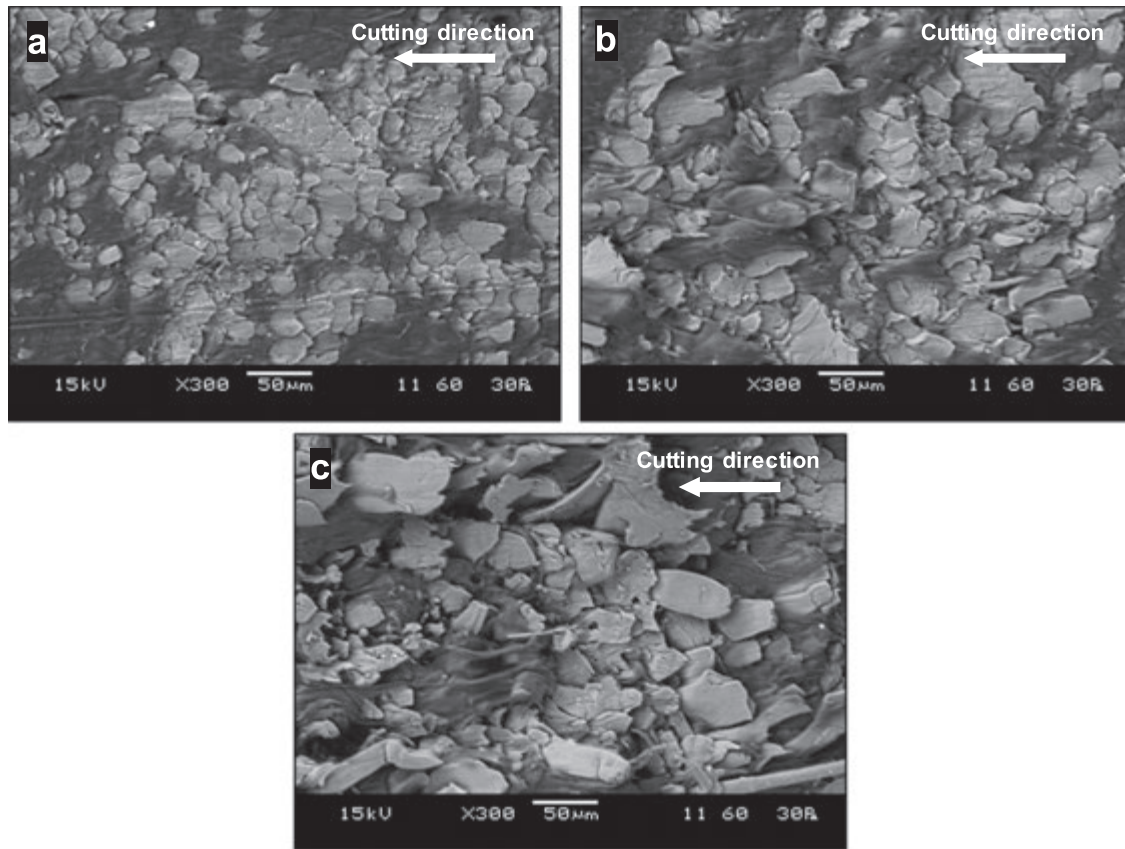


Fig. 22 SEM images of the machined surfaces of UD flax FC with different values of cutting depth. (a) $a_p = 100 \mu\text{m}$, (b) $a_p = 300 \mu\text{m}$, and (c) $a_p = 500 \mu\text{m}$.

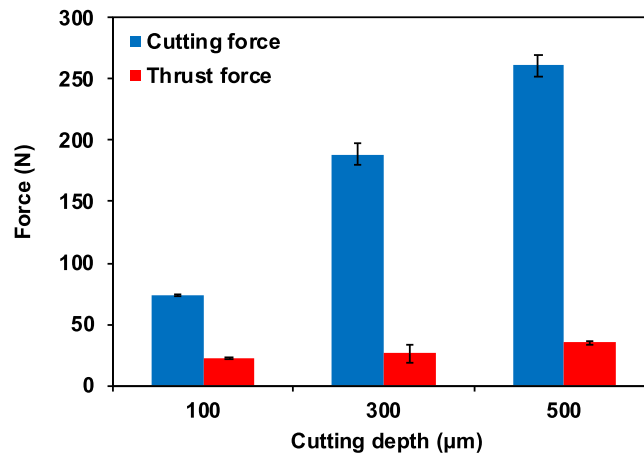


Fig. 23 Machining forces of the orthogonal cutting process of UD flax FC with different values of cutting depth.

Effect of Sample Temperature

The effect of NFC sample temperature on their machinability has been investigated on the same study of rake angle effect presented in Section “Effect of rake angle”. The thermal investigation is performed by comparing the machining of NFC samples at room temperature (25°C) and NFC samples that have been cooled up to 0°C (directly before the tool passes across the sample) using a cooling spray (Chegdani *et al.*, 2018b).

SEM images of Fig. 24 show how lowering the sample temperature before machining influence the cutting behavior of flax fibers. At room sample temperature, flax fibers are deformed toward the cutting direction which makes their shearing difficult.

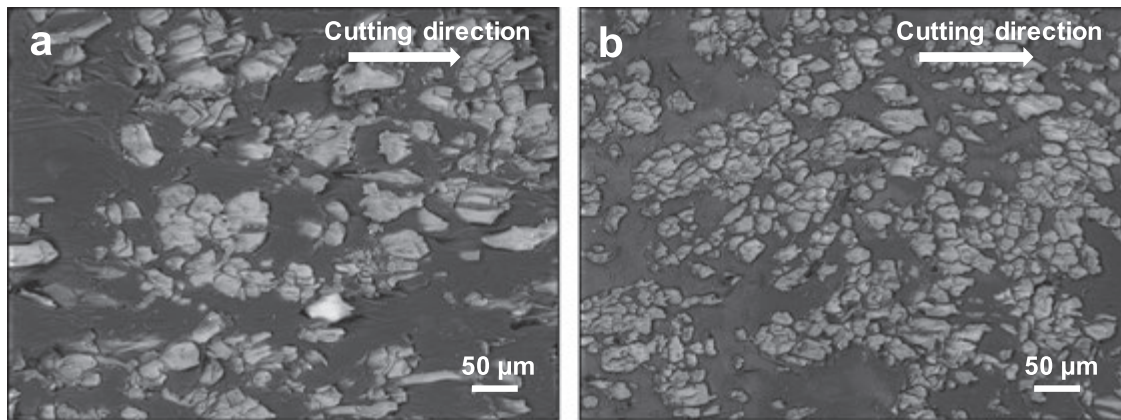


Fig. 24 SEM images of the machined surfaces of UD flax FC at (a) room sample temperature and (b) low sample temperature.

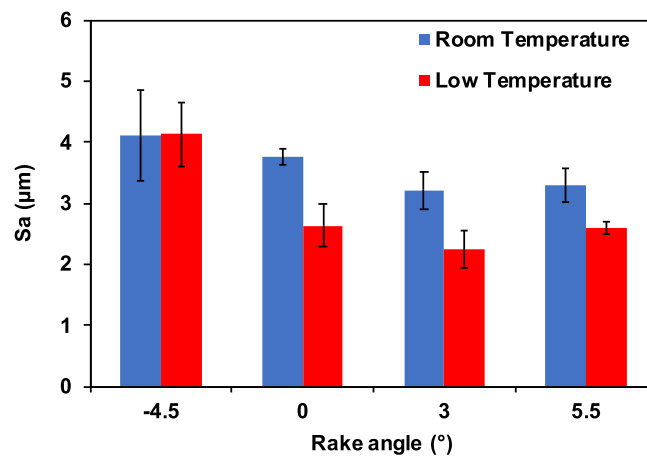


Fig. 25 3D arithmetic mean surface roughness of machined surfaces at room and low sample temperatures.

Therefore, uncut fiber extremities are present on the machining surface in addition to some decohesion zones caused by interfaces damage (Fig. 24(a)). This specific behavior of flax fiber was founded in many previous investigations in this article.

In the case of low sample temperature, cross sections of flax fibers seem to be smaller than that of fibers machined at room temperature (Fig. 24(b)). This shows how much flax fibers are deformed plastically during the cutting operation at room sample temperature. Flax fibers are much better sheared at low sample temperature which avoid the uncut fiber extremities and the high rate of decohesion zones. It can be concluded that lowering the sample temperature increases the cutting contact stiffness at fibers scale with enhances significantly their machinability.

Natural fiber composites considered in this study are manufactured with thermoplastic polymer of polypropylene. The choose of thermoplastic matrices are justified by their reversible transformation which gives to the NFC the ability to be recyclable. Moreover, natural fibers are composed of natural amorphous polymers (hemicellulose and lignin) at nanoscale. Consequently, the polymeric composition on NFC suggests that thermal conditions of machining could have an important impact on the machinability of NFC. Indeed, when decreasing the sample temperature, the rigidity of the polymeric components of the NFC increases and strengthen the fibers stiffness. This gives to the fibers the ability to resist to the transverse deformations caused by the tool crossing.

The thermal effect is also reproduced on the topographic response as shown in Fig. 25. Lowering the sample temperature decreases the surface roughness by improving the shear efficiency of flax fibers in NFC.

Conclusions

Machining of natural fiber composites (NCF) performed with thermoplastic polymers has been investigated in this article. The machinability of NFC is analyzed regarding fibers properties, tool properties, and process parameters. A large exploratory study is presented to cover the main composite structures and the main industrial process applications. The following conclusions can be drawn:

- The removed chip remains continuous at large range of process parameters. This machining behavior of NFC is related to the its ductile behavior generated by the elastoplastic properties of both flax fibers and polymer matrix.
- The cutting behavior of natural fibers is dependent on their mechanical properties. The fiber shearing efficiency of increases by the fiber stiffness increase.
- A fiber orientation of 45° from the cutting direction offers the best fiber shearing and the best machinability of NFC.
- The machinability of NFC is highly dependent on tool geometry. This investigation concludes that the machining tools for NFC should have:
 - (1) Small cutting edge radius which should be less than elementary fiber diameter;
 - (2) Straight cutting flutes with zero helix angle;
 - (3) Positive rake angle.
- Machining of NFC requires the use of:
 - (1) A couple of cutting speed and feed speed that allows the generation of high feed rate: 0.14–0.16 mm/tooth is recommended in this study.
 - (2) Small cutting depths to avoid fiber deformation and interfaces damage: 100 µm id recommended in this study.
- Lowering the sample temperature improves the machinability of NFC by increasing the cutting contact stiffness.

Nevertheless, the majority of these recommendations increases the cutting forces which can be harmful for cutting tools. Moreover, machining with low cutting edge radius means working with uncoated cutting tools which could increase significantly the tool wear. Therefore, deeper investigation should be performed to understand and optimize the wear of cutting tools when machining NFC.

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Multiscale Machinability Analysis of Natural Fiber Composites

Faissal Chegdani, Arts et Metiers Institute of Technology, Mechanics Surfaces and Materials Processing, HESAM University, Châlons-en-Champagne, France

Mohamed El Mansori, Arts et Metiers Institute of Technology, Mechanics Surfaces and Materials Processing, HESAM Université, Châlons-en-Champagne, France and Texas A&M Engineering Experiment Station, Institute for Manufacturing Systems, College Station, TX, United States

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Introduction

The machining behavior of natural fiber composites (NFC) was presented and discussed in authors previous work where it has been demonstrated that the cutting behavior of natural fibers inside the composite is highly sensitive to the variation of material and process parameters (Chegdani *et al.*, 2016, 2015a,b; Chegdani and El Mansori, 2018). Indeed, the machining mechanisms of NFC are more complex because natural fibers are themselves a composite material made of cellulose microfibrils embedded in natural polymers of hemicellulose and pectin (Baley, 2002; Charlet *et al.*, 2007). Thus, the heterogeneity is even present inside each elementary fiber. Moreover, natural fibers are gathered in a bundle of dozen of elementary fibers which have random geometries and diameters (Hossain *et al.*, 2013; Morvan *et al.*, 2003). Therefore, NFRP composites exhibit a multiscale heterogeneous structure from elementary fibers (microscale), then fiber bundles (mesoscale) and finally the overall macroscopic composite material (Bos *et al.*, 2004). Exploring the performances of NFC materials has required a multiscale method either for manufacturing process investigation (Dombia *et al.*, 2015) or for material characterization (Marrot *et al.*, 2014).

For machining processes of NFC parts, a multiscale analysis is also necessary because the mechanical properties of NFC are scale dependent (Chegdani *et al.*, 2017a, 2018c). The standard approaches for the machinability analysis of NFC are not continuously effective to discriminate the effect of some material or process parameters (Chegdani and El Mansori, 2019). In this article, these machinability analysis issues are discussed, and a multiscale approach is proposed to substitute the standard methods for the machinability analysis of NFC.

Machinability Analysis Issues of Natural Fiber Composites

The main issues encountered when analyzing the machinability of NFC are related to the quantification of the surface quality, especially the surface topography of machined surfaces. Indeed, the standard methods for surface topography measurements are not able to discriminate the effects of material/process parameters, even if these effects are revealed on the microscopic observations of the machined surfaces. Fig. 1 gives some examples of these analysis issues.

Fig. 1(a) presents the roughness gain ratio on the machined surfaces of short natural fiber composites to investigate the effect of fiber type and the feed rate on their machinability (Chegdani *et al.*, 2015b). It has been shown in this study that bamboo fiber composites provide the best fiber shearing while miscanthus fiber composites suffer from interfaces damages during the cutting process. Sisal fiber composites generates the worst machinability where an important rate of uncut fiber extremities remains on the machined surface in addition to decohesion zones caused by interfaces damages. These findings were reproduced on the topographic signals. However, the quantification of these topographic signals by the standard method shows that there is no significant difference between the machined surfaces of bamboo FC and Miscanthus FC at the considered range of the feed rate (Fig. 1(a)).

Fig. 1(b) presents the roughness measurements obtained by the standard method on the weft fibers zone (fibers oriented toward the feed direction) for bidirectional flax fiber composites to investigate the effect of the helix angle (H) with the milling process (Chegdani *et al.*, 2016). The microscopic observations have revealed that the weft fibers zones generated random cutting behavior of flax fibers which is dependent on the cutting contact location between the cutting edge and the cross sections of flax fibers. As shown in Fig. 1(b), the standard method for the quantification of the surface roughness at macroscopic scale is not able to discriminate the effect of the helix angle on these machined surfaces.

Moreover, Fig. 1(c) presents the surface roughness of machines surfaces obtained by orthogonal cutting process of unidirectional flax fiber composites to investigate the effect of the cutting depth and the cutting speed (Chegdani and El Mansori, 2018). The microscopic analysis of the machined surfaces reveals that increasing the cutting depth from 100 μm to 500 μm deteriorates the cutting behavior of flax fibers by lowering their shearing efficiency and increasing the interfaces damages. Increasing the cutting speed from 12 m/min to 80 m/min reduces these machining defects. Nevertheless, the quantification of the machined surfaces roughness with the standard method cannot discriminate the effect of the cutting depth at lox cutting speeds as shown in Fig. 1(c).

It can be concluded that the standard methods for the quantification of the surface roughness are not suitable for analyzing the machined surfaces of NFC. The main reason can be related to the contact scale between the NFC and the cutting tool (Chegdani *et al.*, 2015a). Fig. 2 illustrate the multiscale aspect of the cutting contact in the case of NFC materials. At macroscale, the contact is between the cutting edge radius and the global composite structure (Fig. 2(a)). When zooming on the tool/material contact zone, the contact becomes between the fiber bundles and the cutting edge at mesoscale (Fig. 2(b)). A further zoom on the

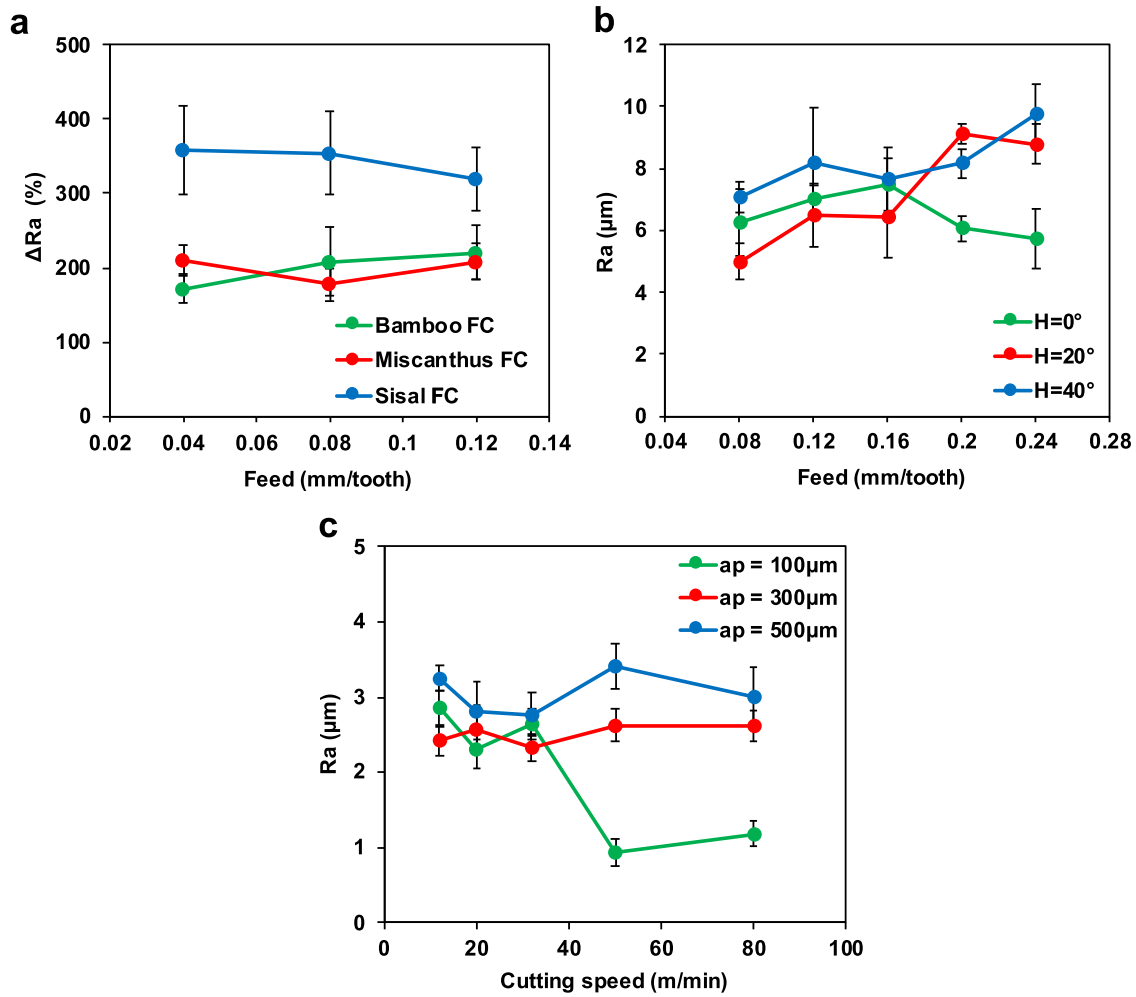


Fig. 1 Surface roughness measurements of machined surfaces of NFC from different investigations. (a) Short fiber composites with milling process, (b) Bidirectional flax fiber composites with milling process, and (c) Unidirectional flax fiber composites with orthogonal cutting process.

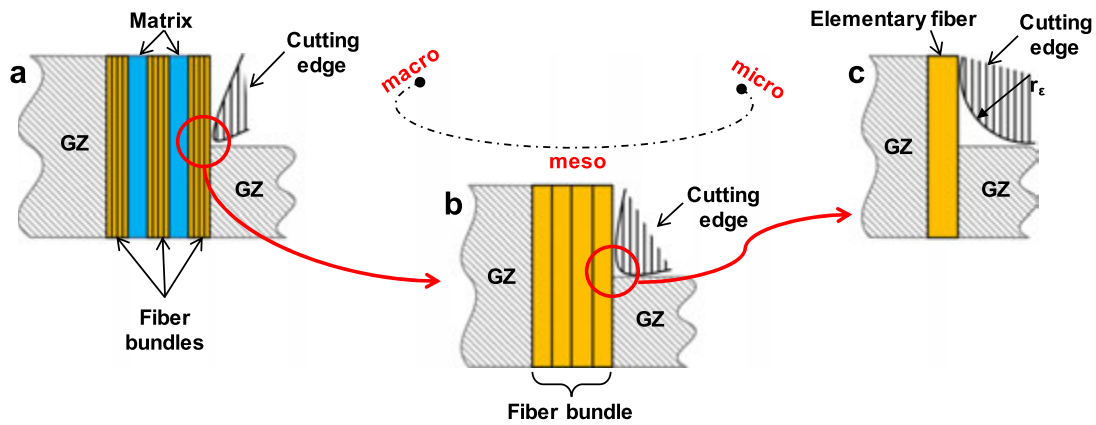


Fig. 2 Schematic illustration of the cutting contact scale at (a) macroscale, (b) mesoscale, and (c) microscale. GZ: Global composite zone, r_e : edge radius.

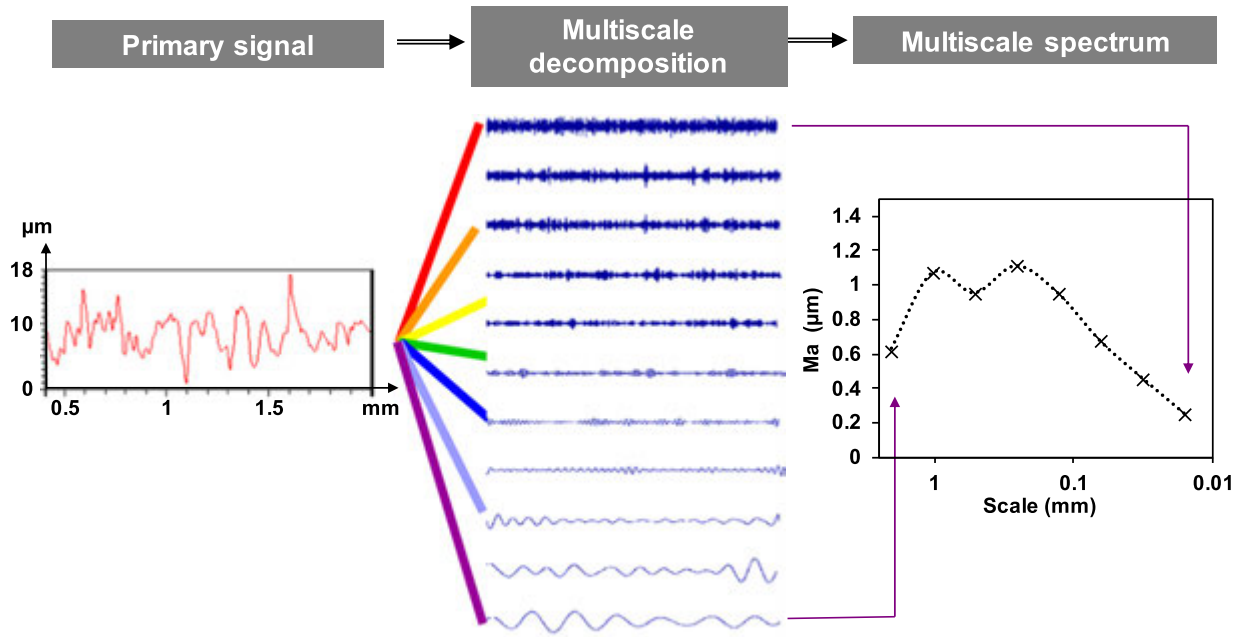


Fig. 3 Schematic illustration of the multiscale decomposition method.

tool/material zone reveals the contact at microscale between the cutting edge and the elementary fiber as shown in Fig. 2(c). Therefore, A multiscale method for machined surface characterization should be adopted to analyze the machinability of NFC. The proposed multiscale method is presented and discussed in next sections.

Multiscale Analysis Methodology

The multiscale method is based on a multiscale decomposition of topographic signals using wavelet transform. The idea is to find at which range, each material/process variable affects the morphology of the machined surface. The wavelet transform can be used by discrete wavelet transform (DWT) (Chegdani *et al.*, 2017b) or continuous wavelet transform (CWT) (Chegdani *et al.*, 2016). As described in Fig. 3, the aim of this wavelet transform is to decompose the primary topographic signal obtained by the standard method through a series of high-pass and low-pass filters to analyze the high and low frequencies (Chen *et al.*, 2012; Chowdhury *et al.*, 2013; Dick *et al.*, 2012; Katunin, 2011; Peng *et al.*, 2009; Qiu *et al.*, 2016). Since the high frequencies correspond to the micro-roughness and the low frequencies correspond to the waviness, the wavelet transform can quantify the surface morphology at different scale levels.

Discrete Wavelet Transform (DWT)

The basic functions to filter the primary topographic signal in DWT are obtained from a single prototype wavelet called the “Mother” wavelet ($\psi(x)$) by translation and dilation (Chen *et al.*, 1995; Daubechies, 1992). The mother wavelet is discretized using the Eq. (1) where m and n are, respectively, the translation and dilation parameters. Then, the logarithmic scaling of both dilation and translation steps ($a_0 = 2$ and $b_0 = 1$) generates an orthogonal wavelet shown in Eq. (2). The DWT of the global topographic signal (called $f(x)$) is defined by the Eq. (3) where $\bar{\psi}_{m,n}(x)$ is the conjugate of the wavelet function. Finally, the reconstruction of the global topographic signal $f(x)$ is given by the Eq. (4).

$$\psi_{m,n}(x) = \frac{1}{\sqrt{a_0^m}} \psi\left(\frac{x - nb_0 a_0^m}{a_0^m}\right) \quad (1)$$

$$\psi_{m,n}(x) = 2^{-m/2} \psi(2^{-m}x - n) \quad (2)$$

$$W(m, n) = \langle \bar{\psi}_{m,n}(x), f(x) \rangle \quad (3)$$

$$f(x) = \sum_{m,n} W(m, n) \psi_{m,n}(x) \quad (4)$$

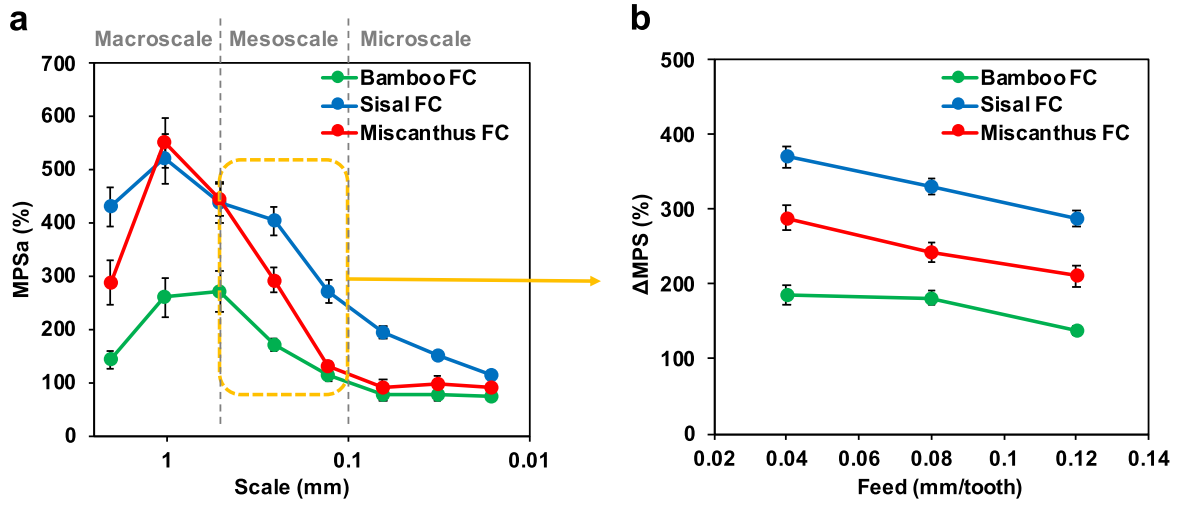


Fig. 4 (a) Multiscale process signature on machined surfaces of short fiber composites obtained by discrete wavelet transform. (b) Process signature at mesoscale on machined surfaces of short fiber composites.

Continuous Wavelet Transform (CWT)

The continuous wavelet transform of the initial topography signal $f(x)$ is defined by the Eq. (5) where $\psi_{a,b,\theta}(\vec{x}) = \psi_{a,b}(r_{-\theta}(\vec{x}))$ and r_{θ} is the rotation operator defined by the Eq. (6). For 2D continuous wavelet transform, $x = (x, y)$.

In Eq. (5), “ a ” is the contraction coefficient, $\vec{b} = (b_x, b_y)$ the translation coefficient in the x and y directions. Therefore, each component altitude of the global topography signal “ f ” at the scale “ i ” in the “ (x, y) ” point coordinate $f_i(\vec{x}, \theta)$ can be thus obtained in each analysis direction “ θ ” by inverse wavelets transform.

$$W_{\psi}^f(a, b, \theta) = \frac{1}{a} \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} f(\vec{x}) \psi_{a,b,\theta}^*(\vec{x}) d\vec{x} \quad (5)$$

$$r_{\theta}(x, y) = (x \cos \theta + y \sin \theta - x \sin \theta + y \cos \theta) \quad (6)$$

Multiscale Roughness Quantification

After the wavelet transform, the arithmetic mean roughness can be quantified at each scale of the decomposition. This multiscale arithmetic mean roughness is called “ Ma ” for 1D line topographic profiles (x) and “ SMa ” for 2D surface topographic profiles (x, y). This arithmetic mean value can be obtained using the Eqs. (7) and (8) where $f_i(x)$ and $f_i(x, y)$ are respectively the component altitude of the global topographic signal at the scale “ i ” in the point coordinate (x) and (x, y). “ N ” and “ M ” represent respectively the number of points in the x and the y directions (Chegdani and El Mansori, 2019).

$$M_a(i) = \sum_{x=1}^M \frac{|f_i(x)|}{M} \quad (7)$$

$$SM_a(i) = \sum_{x=1}^M \sum_{y=1}^N \frac{|f_i(x, y)|}{MN} \quad (8)$$

Finally, the obtained values of Eq. (7) or Eq. (8) allow the determination of the multiscale process signature (MPS) which is equivalent to a transfer function that can be compared directly with the multiscale modifications of the surface topography. This MPS depicts the signatures of the finishing process in terms of essential changes of the surface state produced on the original surface (El Mansori et al., 2010). The MPS is defined at each scale “ i ” by Eqs. (9) and (10) for 1D line topographic profiles and 2D surface topographic profiles, respectively. In these two equations, “ F ” and “ I ” refer to final state and the initial state of the machined surfaces, respectively.

$$MPS(i) = \frac{M_a^F(i) - M_a^I(i)}{M_a^I(i)} \times 100 \quad (9)$$

$$MPS(i) = \frac{SM_a^F(i) - SM_a^I(i)}{SM_a^I(i)} \times 100 \quad (10)$$

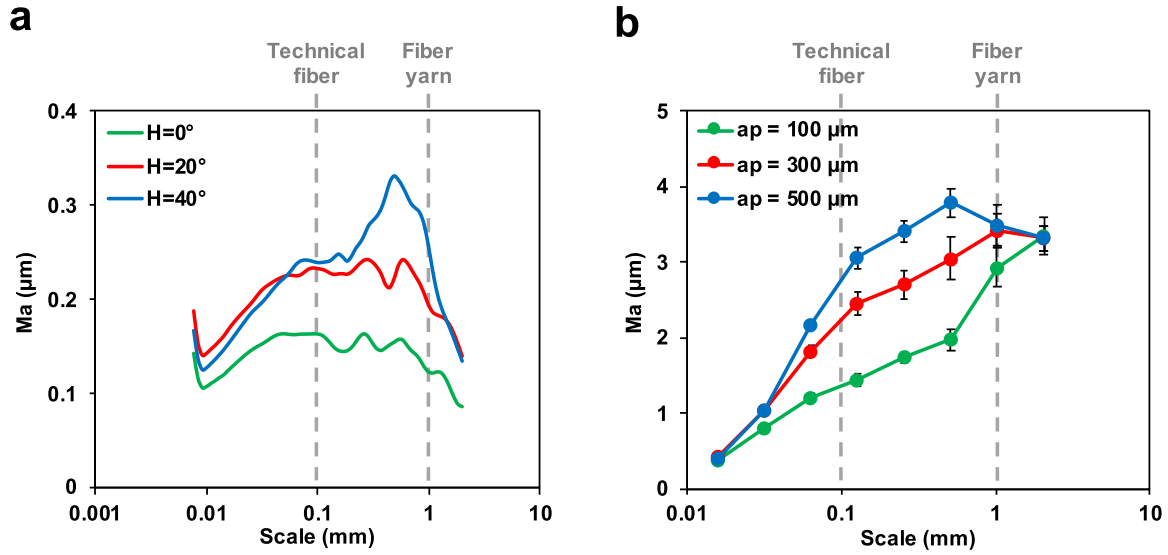


Fig. 5 (a) Multiscale process signature on machined surfaces of bidirectional flax fiber composites obtained by continuous wavelet transform. (b) Multiscale process signature on machined surfaces of unidirectional flax fiber composites obtained by discrete wavelet transform.

After filtering the pertinent scales by the *MPS*, the mean process signature (ΔMPS) can be calculated to assess the morphology modifications due to the machining process without the parasitic effects of the impertinent scales. ΔMPS is defined by Eq. (11) where “*P*” is the number of the pertinent scales in the wavelet decomposition.

$$\Delta MPS = \sum_{i=1}^P \frac{MPS(i)}{P} \quad (11)$$

Machinability Analysis of Natural Fiber Composites With the Multiscale Methodology

Scale Effect on the Machined Surface Roughness

The machining analysis issues presented in Section “Machinability Analysis Issues of Natural Fiber Composites” have been handled by the multiscale method described in Section “Multiscale Analysis Methodology”. Each primary signal obtained by the standard methods has been processed using either the DWT or the CWT.

Fig. 4(a) shows the multiscale spectrum obtained from the primary signals of the machined surface of short fiber composites investigated in (Chegdani *et al.*, 2015b). The scale effect of the topographic response of the machines surfaces is clearly obvious. The roughness is at its lowest values at microscale and increases by increasing the analysis scale until reaching a macroscale of 1 mm. The roughness decreases after exceeding this macroscale value.

Fig. 4(a) show also that the effect of fiber type is clearly discriminated at the scale range between 0.1 mm and 0.5 mm. this mesoscale range correspond to the size of the fiber bundles used in the short fiber composites materials. Therefore, the relevant scale of the machinability analysis of NFC is related to the mesoscale of the natural fibrous reinforcement.

By considering only the mesoscale for the analysis of the machined surface roughness, Fig. 4(b) reveals that the effects of fiber type and cutting feed are obviously distinguished where increasing the cutting feed decreases the surface roughness. Moreover, Bamboo fibers, that have the highest rigidity, generate the lowest surface roughness while sisal fibers, that have the lowest rigidity, generate the highest surface roughness. These findings shown in Fig. 4(b) are not provided on the results of the standard analysis methods presented in Fig. 1(a) where the effect of fiber type is not differentiated between bamboo and miscanthus fibers.

Fig. 5(a) shows the multiscale spectrum obtained from the primary signals of the machined surface of bidirectional flax fiber composites investigated in (Chegdani *et al.*, 2016). The spectrums in Fig. 5(a) were obtained as the mean of 20 topographic profiles extracted from both warp fiber zones and weft fiber zones in order to generate a representative output for all the machined surface.

It can be seen from Fig. 5(a) that the effect of tool helix angle is revealed at the scale range between 0.1 mm and 1 mm. This scale range matches to the flax reinforcement structure size used in the considered composite because 0.1 mm corresponds approximately to the diameter of the technical fiber and 1 mm corresponds approximately to the diameter of the flax yarns formed by the technical fibers.

Fig. 5(b) shows the multiscale spectrum obtained from the primary signals of the machined surface of unidirectional flax fiber composites investigated in (Chegdani and El Mansori, 2018) with a cutting speed of 20 m/min. In this multiscale study, the analysis scale affects also the roughness response that increases by increasing the analysis scale. As for the case of bidirectional flax fiber composites (Fig. 5(a)), the effect of cutting depth is revealed at the scale range between 0.1 mm and 1 mm which correspond to the flax reinforcement structure size.

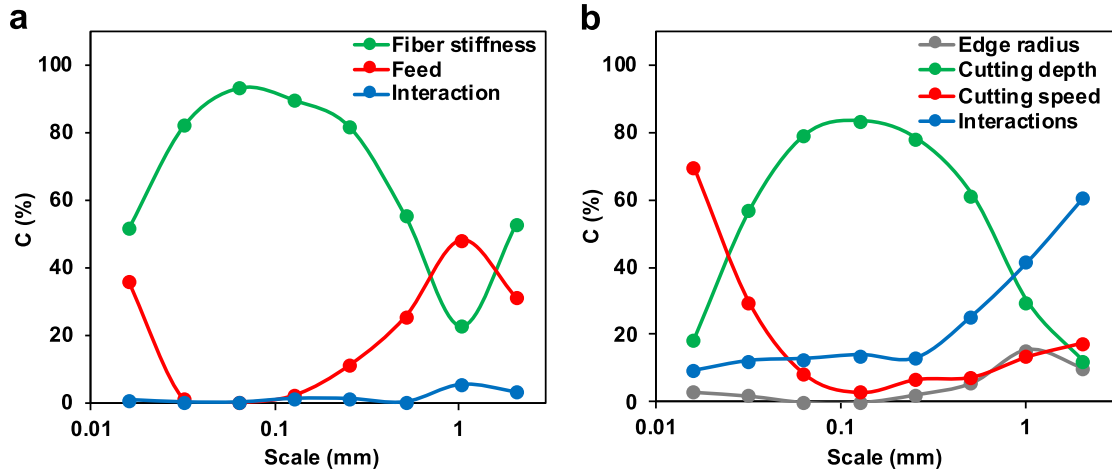


Fig. 6 Contribution rate of process parameters for (a) milled surfaces of short fiber composites, and (b) machined surfaces of unidirectional flax fiber composites with orthogonal cutting process.

Multiscale Contribution of Machining Parameters on the Surface Roughness

To quantify the effect of source factors on the roughness level at each scale of the decomposition, mathematical models for machining of NFC using regression analysis and the analysis of variance (ANOVA) (Davim and Reis, 2005; Gonzalez *et al.*, 2015) were elaborated. Linear regression analysis for the results of the multiscale surface roughness analysis was considered.

Sequential approach or Type I sum of squares was used to test main effects and interaction behaviors in ANOVA. It allows, first, to allocate the part of the explained variance to the main effects (one after the other), then to the two-way interaction(s) (one after the other) and then to increasingly higher-order interactions if present (Kherad-Pajouh and Renaud, 2010).

Experimental and predicted multiscale surface roughness responses are compared. The sum of squares (SS) and the fitted residual sum of squares (RSS) of these gaps are computed and collected from all the parallel models to form the predictive residual sum of squares (PRESS) which estimates the predictive ability of the model. The goodness-of-fit is evaluated for the considered model with the measure of the squared correlation coefficient (R^2) and the cross-validated squared correlation coefficient (Q^2) (Wold *et al.*, 2001). Where:

$$R^2 = 1 - \frac{RSS}{SS} \quad (12)$$

$$Q^2 = 1 - \frac{PRESS}{SS} \quad (13)$$

R^2 is a real number between zero and one. A large value of R^2 indicates a better fitness of the model to the data. The predictive capability of a model is generally determined by Q^2 which is usually between zero and one. A higher Q^2 value indicates a more reliable model with excellent predictive power (Wang *et al.*, 2013). Q^2 can be negative for very poor models.

After the validation of the model with R^2 and Q^2 , F -test (Massart *et al.*, 1998) was used to quantify the significance of each input working factor " α " by Eq. (14) where MS_{reg} is the mean square due to regression and MS_r is the residual mean square.

$$F(\alpha) = \frac{MS_{reg}(\alpha)}{MS_r} \quad (14)$$

To quantify the contribution of the studied material/process parameters, ANOVA of input variables influence has been performed at each multiscale response of surface quality (i.e., at each scale " a " of the decomposition) using XLSTAT software. The contribution ratio of each " α " factor (C_α) was calculated with the Fisher criterion test ($F(\alpha)$) at the correlation coefficient (R^2) confidence (Davim and Reis, 2005; Massart *et al.*, 1998). Eq. (15) defines the contribution ratio of each factor.

$$C_\alpha = \frac{F(\alpha)}{\sum_\alpha F(\alpha) \times R^2} \quad (15)$$

The ANOVA analysis was applied to the multiscale roughness data shown in Fig. 4(a) where the contribution rates of fiber type and cutting feed were determined (Chegdani *et al.*, 2017b). Fig. 6(a) reveals that the contribution rates of natural fiber stiffness and feed factors are different at each analysis scale and present specific behaviors at three characteristic zones that are closed to the characteristic zones defined by multiscale surface roughness analysis of Fig. 4(a). At microscale, fiber stiffness contribution increases significantly by scale increasing while feed contribution decreases significantly until becoming negligible. Interaction contribution is insignificant in this zone and also at mesoscale. This mesoscale area reveals an opposite behavior comparing to the microscopic zone where fiber stiffness contribution decreases significantly by scale increasing while feed contribution increases by scale increasing. This trend is spread until the scale $a = 1$ mm that shows the most significant contribution of the interactions between fiber stiffness and feed effects. Feed effect reaches its maximum and fiber stiffness effect reaches its minimum. At

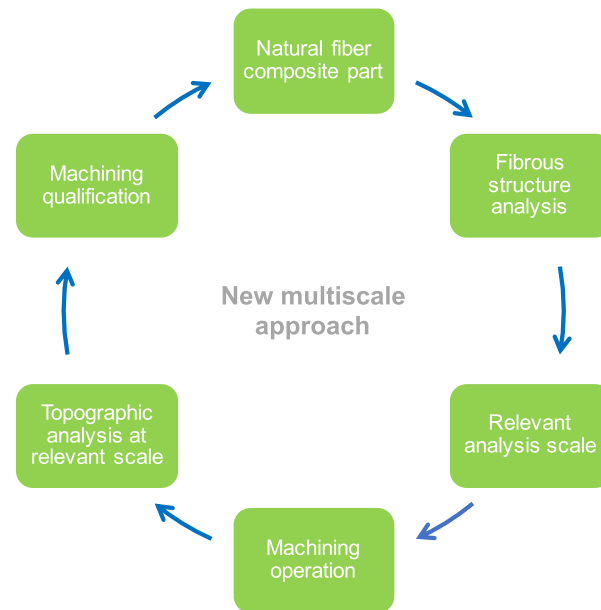


Fig. 7 Principle of the multiscale approach for machinability analysis of natural fiber composites.

macroscale, fiber stiffness contribution re-increases by scale increasing while feed contribution re-decreases by scale increasing. This multiscale behavior of the contribution rates of material/process parameters is intrinsically related to the multiscale structure of the NFC materials (Chegdani *et al.*, 2017b).

The ANOVA analysis was also applied to the multiscale roughness data shown in Fig. 5(b) in order to determine the contributions of cutting depth, cutting speed and edge radius (Chegdani and El Mansori, 2018). Fig. 6(b) presents the contribution rate of each process parameter at each analysis scale. It can be seen the low contribution of the cutting edge radius regarding the other parameters until the macroscale. The behavior of the multiscale contribution rates confirms the previous results shown in Fig. 5(b). Indeed, Fig. 6(b) shows the significant impact of the cutting depth at the relevant scale identified in Fig. 5(b). At macroscopic scale, the random distribution of technical fibers influences the contribution of the process parameters and increases the contribution of the interactions (Chegdani and El Mansori, 2018).

New Multiscale Approach for the Machinability Analysis of Natural Fiber Composites

Principle of the New Multiscale Approach

Based on the results presented in Section “Machinability Analysis of Natural Fiber Composites With the Multiscale Methodology”, machining qualification of natural fiber composites requires the selection of the relevant scale for the cutting process analysis. The pertinent scale corresponds to the size of the fibrous structure regardless of the reinforcement structure type (Chegdani and El Mansori, 2019). This specific analysis allows an efficient discrimination of the effects of material/process parameters.

Concretely, Fig. 7 illustrates the principle of the new multiscale approach that should be applied for the analysis of the machinability of NFC materials. Indeed, to qualify effectively the machinability of the NFC parts, the natural fibrous structure should be first analyzed in order to determine the fibrous structure size. The value of the fibrous structure size will correspond then to the relevant analysis scale on which the topographic analysis of the machined surfaces of NFC parts should be performed to evaluate their machinability.

The relationship between the relevant scale and the fibrous structure size has been previously confirmed by nanoindentation and scratch test measurements that show the scale effect on the tribo-mechanical performances of flax fibers (Chegdani *et al.*, 2017a, 2018c). Unlike glass fibers that have a homogeneous mechanical behavior, the tribo-mechanical response of flax fibers induces a multiscale behavior that depends on the mechanical contact scale. This reveals the specificity when cutting natural fibers and the importance of considering the analysis scale for robust machining analysis of NFC materials in industrial applications.

Validation of the New Multiscale Approach

The validation of the new multiscale approach for machinability analysis of NFC has been performed on an industrial part provided by Faurecia Automotive Industry (Chegdani and El Mansori, 2019). The industrial NFC is a sandwich part as shown in Fig. 8(a). The skins are composite plates molded of unidirectional flax fibers and Acrodur bio-resin. The honeycomb structure is made of cardboard. Each skin of the sandwich NFC consists of three layers of unidirectional long flax fibers bonded by the Acrodur

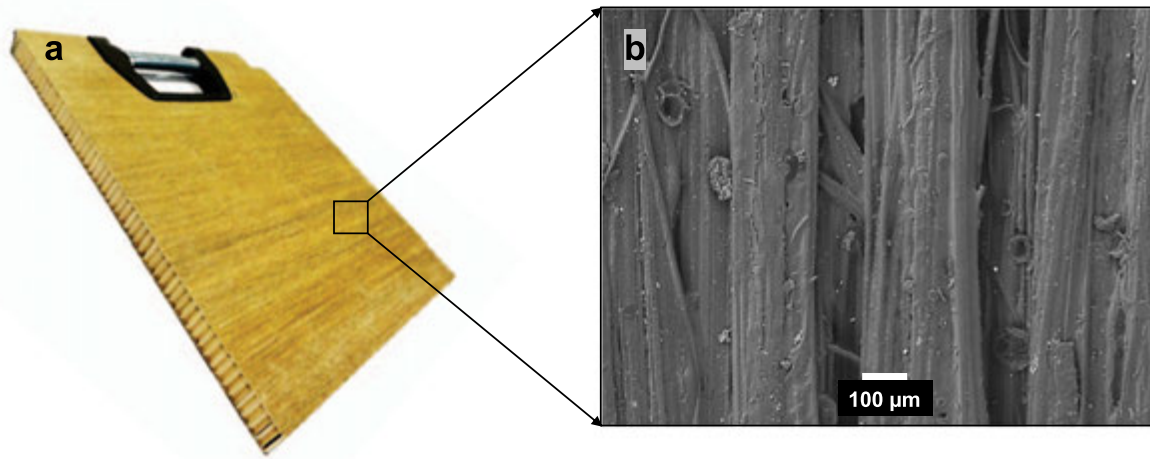


Fig. 8 (a) Image of the industrial sandwich part. (a) SEM image of the flax fibrous reinforcement in the sandwich skins.

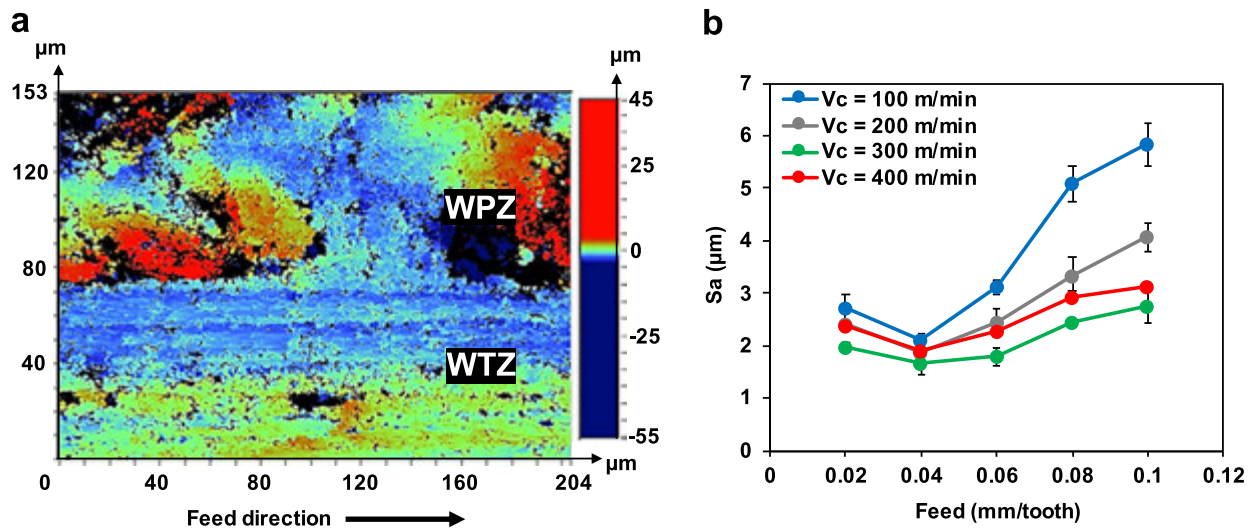


Fig. 9 (a) topographic image at the relevant scale on the machined surface of the sandwich skin. (b) Arithmetic mean surface roughness of the machined surface of the sandwich skin performed at the relevant scale.

resin. The density of the fibers in each layer is about 200 g/m^2 . This gives the skin a thickness of 1 mm. The three layers of UD flax/Acrodur in each skin have a fiber orientation configuration of $0^\circ/90^\circ/0^\circ$. This fibrous reinforcement structure will generate warp fiber zone (WPZ) and weft fiber zone (WTZ) as for the case of bidirectional flax fiber composites investigated in Section “Machinability Analysis Issues of Natural Fiber Composites” and Section “Machinability Analysis of Natural Fiber Composites With the Multiscale Methodology”. The thickness of the cardboard honeycomb structure is 20 mm.

As suggested by the new multiscale approach, the fibrous reinforcement is analyzed with SEM observation as shown in Fig. 8(b). It can be seen that the fibrous structure is in form of non-twisted technical flax fibers that have diameter values between 150 μm and 200 μm.

Milling process is considered to perform the machining experiments on the sandwich part. The analysis focuses on the profile surface state of the sandwich skin (UD flax and Acrodur resin). The behavior of the cardboard honeycomb structure is not considered. The effect of cutting speed and cutting feed is investigated in this study. The topographic analysis of the machined surfaces of the sandwich skins are realized with respect to the recommendations of the multiscale approach. Indeed, the optical objective of the interferometer has been chosen and adjusted to produce topographic image dimensions that correspond to the scale of the technical fibers size ($\sim 150\text{--}200 \text{ μm}$).

Fig. 9(a) present a typical topographic image of the machined surfaces at the relevant scale that allows to cover the two fiber zones (WPZ and WTZ). The resulting arithmetic mean roughness of the machined surfaces at the relevant scale are presented in

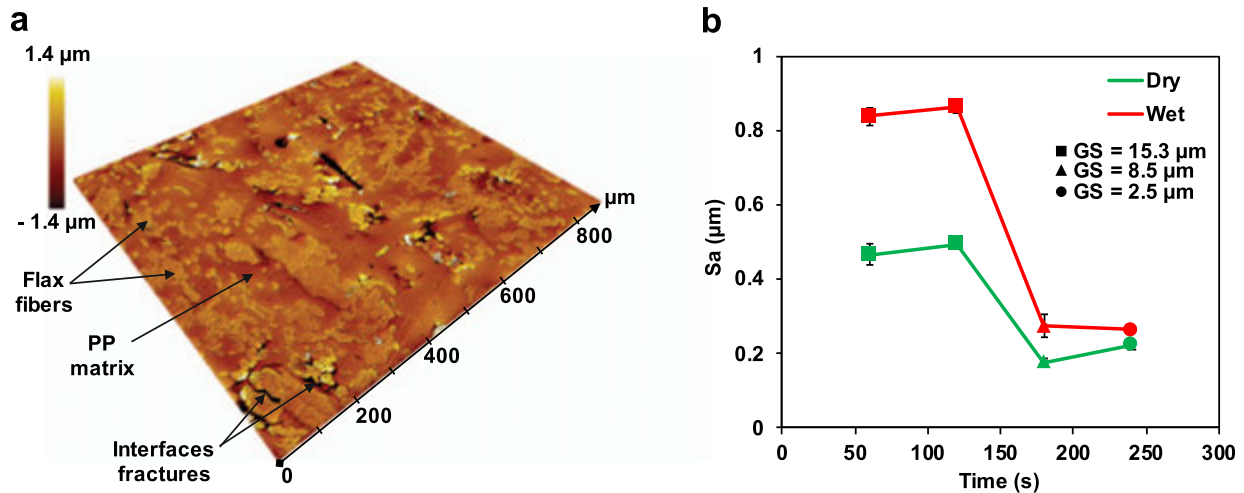


Fig. 10 (a) typical topographic image of the polished surfaces of unidirectional flax fiber composites at the relevant scale. (b) Arithmetic mean surface roughness of the polished surfaces of unidirectional flax fiber composites at the relevant scale.

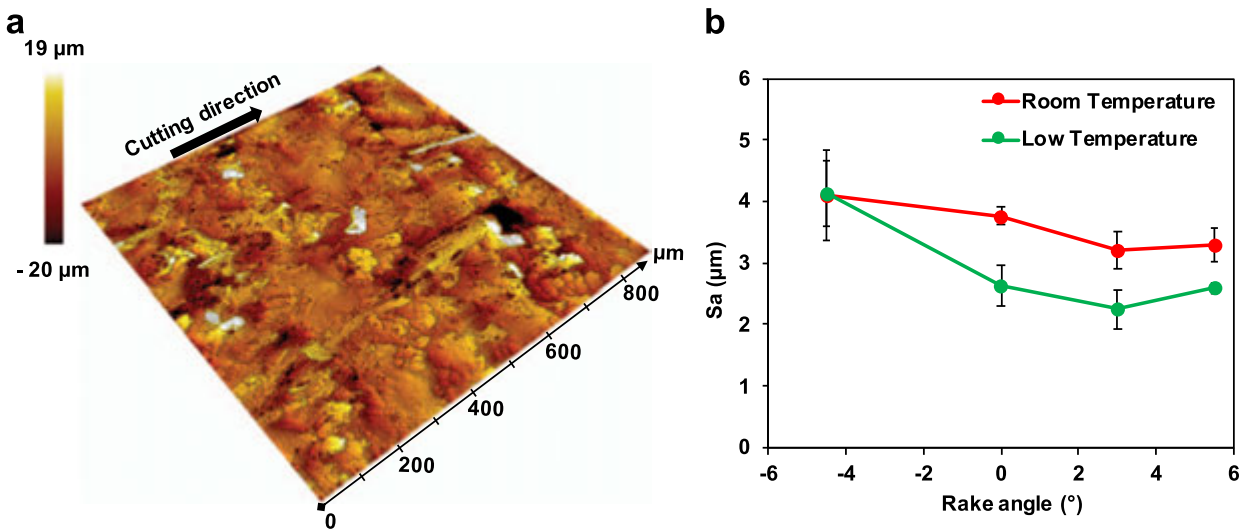


Fig. 11 (a) typical topographic image of the machined surfaces of unidirectional flax fiber composites at the relevant scale. (b) Arithmetic mean surface roughness of the machined surfaces of unidirectional flax fiber composites at the relevant scale.

Fig. 9(b). Unlike the standard topographic analysis on bidirectional flax fiber composites that shows a random behavior of the surface roughness results (**Fig. 1(b)**), the topographic analysis performed in accordance with the new multiscale approach allow to get a pertinent evaluation of the effect of both the cutting speed and the cutting feed as shown in **Fig. 9(b)**. Overall, the surface roughness increases by increasing the feed rate and decreases by increasing the cutting speed. The multiscale analysis approach leads to find an optimum of cutting speed and feed rate for machining the industrial sandwich part. The roughness is at its lowest value for feed rate of 0.04 mm/tooth and for a cutting speed of 300 m/min. Consequently, these cutting conditions are the best-suited parameters for this material to guarantee the smoothest surfaces after machining with fewer surface damages.

The new multiscale approach has also been validated during the investigation of the surface forming of unidirectional flax fibers reinforced polypropylene composites using the mechanical polishing process at dry and wet polishing conditions (**Chegdani et al., 2018a**). As shown in Section "Scale Effect on the Machined Surface Roughness" and **Fig. 5(b)**, the relevant scale for this kind of NFC is in the scale range between 0.1 mm and 1 mm which correspond to the flax reinforcement structure size. Therefore, the topographic analysis of the polished surfaces has been performed using an optical objective of the interferometer that generates a topographic image size of $840 \times 840 \mu\text{m}^2$ as shown in **Fig. 10(a)**. The resulting arithmetic mean roughness of the polished surfaces at the relevant scale are presented in **Fig. 10(b)**. The roughness calculation at the relevant scale allows a good discrimination of the effect of each polishing configuration. Indeed, wet polishing induces more surface roughness than dry polishing because of the fracture of the interfaces caused by water lubrication (**Chegdani et al., 2018a**). These interface fractures are more

intense when polishing with high grit size (GS in Fig. 10(b)) which further increases the surface roughness of the wet polished surfaces comparing to the dry polished surfaces.

The same unidirectional flax fiber composites have been used in other study to investigate the effects of sample temperature and rake angle on the machining behavior of the NFC using the orthogonal cutting process (Chegdani *et al.*, 2018b). As for the investigation of the polishing process, the topographic analysis of the machined surfaces is performed using an optical objective of the interferometer that generates a topographic image size of $840 \times 840 \mu\text{m}^2$ as shown in Fig. 11(a). The resulting arithmetic mean roughness of the machined surfaces at the relevant scale are presented in Fig. 11(b). It can be seen once again that the roughness calculation at the relevant scale allows a good discrimination of the effect of sample temperature and rake angle for the orthogonal cutting process of unidirectional flax fiber composites. Lowering the sample temperature leads to decrease the machined surface roughness by increasing the rigidity of flax fibers and polymer matrix (Chegdani *et al.*, 2018b). This investigation with the new multiscale approach suggests also to perform the cutting operation with positive rake angle to increase the cutting contact stiffness and then reduce machined surface roughness as shown in Fig. 11(b).

Conclusions

Machining of natural fiber composites encounters some issues related to the machinability analysis. Indeed, the qualification of the machinability of natural fiber composites is complicated with the standard approach, especially for the quantification of the machined surfaces because of the complex multiscale structure of natural fiber composites. In this study, these machinability analysis issues are presented and discussed. Moreover, a new multiscale approach is developed and proposed to address the machinability qualification of natural fiber composites. The main following conclusions can be drawn:

- Regardless of the natural reinforcement structure types, the relevant scale for analyzing the machined surfaces of natural fiber composites are the scale that corresponds to the fibrous structure size.
- Unlike the standard approaches for the quantification of the machined surface roughness, the new multiscale approach recommends analyzing first the natural fibrous structure and determine its size. The fibrous structure size value should correspond to the pertinent scale on which the topographic analysis of machined surfaces should be performed. This will allow an efficient discrimination of the material/process effects on the surface quality of natural fiber composites.
- The new multiscale approach has been validated on different machining investigations and has shown its strength to provide an appropriate machinability qualification of natural fiber composites.

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Numerical Modeling of the Machining Behavior of Natural Fiber Composites

Faissal Chegdani, Arts et Metiers Institute of Technology, Mechanics Surfaces and Materials Processing, HESAM University, Châlons-en-Champagne, France

Mohamed El Mansori, Arts et Metiers Institute of Technology, Mechanics Surfaces and Materials Processing, HESAM Université, Châlons-en-Champagne, France and Texas A&M Engineering Experiment Station, Institute for Manufacturing Systems, College Station, TX, United States

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Introduction

Machining of natural fiber composites (NFC) starts arousing the interest of many researchers due to the increase in the use of NFC materials in different industrial applications (Lotfi *et al.*, 2019; Nassar *et al.*, 2017; Rajmohan *et al.*, 2019; Roy Choudhury *et al.*, 2018; Vinayagamoorthy and Rajmohan, 2018). The machinability of NFC has shown different technical issues related to the well-known complex multiscale cellulosic structure of natural fibers (Baley, 2002). Indeed, the machining behavior of NFC is highly sensitive to the small variation of material and process parameters (Chegdani *et al.*, 2018a, 2016, 2015a,b; Chegdani and El Mansori, 2018; Chegdani and Mansori, 2018). Moreover, a new multiscale approach has been developed to perform the machinability qualification of NFC materials. This approach postulates that the machining investigation of NFC requires the selection of the pertinent analysis scale that corresponds to the size of the natural fibrous reinforcement structure (Chegdani and El Mansori, 2019).

Despite the effectiveness of the new machinability qualification approach for NFC, its application presents some issues related to the high variability of natural fibers in terms of shape and diameter (Hossain *et al.*, 2013). Therefore, the new multiscale approach should be adapted and re-applied to each fibers harvest because, depending on the environmental conditions of growth, the fibrous structure size could vary significantly which is time and cost consuming for the experimental machinability qualification.

The use of the numerical technologies could be an efficient way to resolve these issues. Indeed, developing a numerical model for machining of NFC could reduce considerably the time and the cost of the machinability analysis during the experimental validation of new NFC products. For this aim, this article proposes a 2D micromechanical finite element model to simulate the orthogonal cutting behavior of NFC at the pertinent scale of natural fibers.

Finite element (FE) method has previously been applied to model the machining behavior of synthetic fiber composites at microscale (Dandekar and Shin, 2012, 2008; Gao *et al.*, 2016; Rao *et al.*, 2007a,b). All the FE models are developed for synthetic fiber reinforced thermoset composites because of their heavy use in structural industrial applications. Synthetic fibers have been modeled with an elastic behavior and brittle failure based on the maximum failure stress criterion. Polymer matrices have been modeled with an elasto-plastic behavior and a ductile damage. Interfaces have been usually modeled with the cohesive zone model (CZM) while some researchers consider the interfaces as solid continuum elements (Gao *et al.*, 2016).

For NFC materials, natural fibers cannot be modeled using the maximum failure stress criterion at microscale. This is because natural fibers have a cellulosic structure that induces a visco-elasto-plastic behavior with both tensile tests (Charlet *et al.*, 2007; Placet *et al.*, 2014) and nanoindentation experiments (Chegdani *et al.*, 2018b, 2017; Keryvin *et al.*, 2015). Also, the well-known Johnson-cook model for ductile materials (Johnson and Cook, 1985) cannot be applied to natural fibers because it assumes an isotropic hardening of the considered material (Abaqus Analysis User's Manual, 2011), while natural fibers show an anisotropic behavior (Baley, 2002).

In this article, the FE machining model will be developed for natural fibers reinforced thermoplastic composites that are the most used in the industry to insure the recyclability of these eco-friendly materials.

Finite Element Modeling Strategy

Finite Element Model Setup

Fig. 1 shows the geometrical setup of the FE model carried out using ABAQUS/Explicit software (version 6.11–2) (Abaqus Analysis User's Manual, 2011). The microscopic-based model consists of a bundle of four elementary flax fibers embedded in the polymer matrix. The interfaces between flax fibers and the matrix are modeled with cohesive elements that have a thickness of 1 μm . Each elementary fiber has a diameter of 15 μm . The cutting tool is considered as an analytical rigid body where the movement is controlled by a reference point. Flax fibers are oriented with an angle " θ " from the cutting direction. A plane stress analysis, which is more suitable for FEM cutting of composites (Lasri *et al.*, 2009), is considered in this numerical investigation. Both flax fibers and PP matrix are meshed with 4-node bilinear plane stress quadrilateral elements (CPS4R) and 2 μm of mesh size. Cohesive interfaces are meshed with a 4-node two-dimensional cohesive element (COH2D4) and 1 μm of mesh size.

As for synthetic fiber composites, the thermal effect is not considered in this micromechanical study because the heat generation in orthogonal cutting requires a large cutting length (Nayak *et al.*, 2004; Santiuste *et al.*, 2010). The cutting length in the current model does not exceed 200 μm .

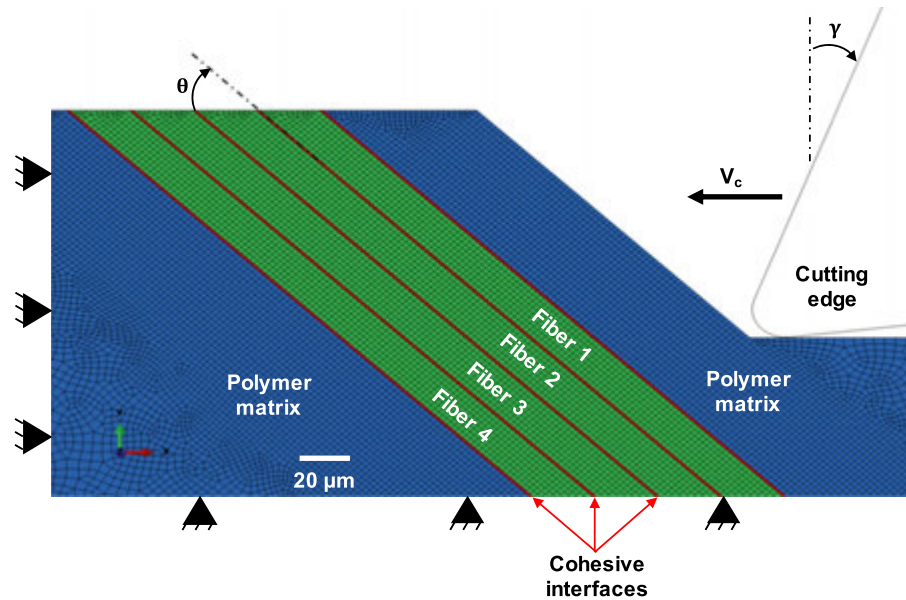


Fig. 1 Geometrical setup of the micromechanical EF model for machining NFC.

Two NFC materials are considered in this article to model the machining behavior:

- Unidirectional flax fiber reinforced polypropylene composite (Flax/PP);
- Unidirectional flax fiber reinforced polylactic-acid composite (Flax/PLA).

Next section will describe the modeling laws used for each composite component.

Finite Element Modeling of Natural Fibers

Unlike synthetic fibers such as glass and carbon, natural fibers exhibit an anisotropic behavior due to their cellulosic structure as shown in **Fig. 2(a)** where natural fibers are structured as a stacking of cell walls and each cell wall is itself a composite material of cellulose microfibrils embedded in natural amorphous polymers of hemicellulose, pectin, and lignin (Baley, 2002; Charlet *et al.*, 2007). The major cell wall that controls the fiber behavior is “S2” because of its predominant volume. In the cell wall S2, the cellulose microfibrils are oriented with a small angle called the microfibrillar angle (MFA) (Baley, 2002).

This cellulosic structure of natural fibers induces an orthotropic behavior and the mechanical behavior of the elementary flax fiber depends on the cellulose microfibrils orientation toward the fiber axis. Indeed, tensile tests on elementary flax fibers reveal a non-linearity as shown in the stress/strain curve of **Fig. 2(b)** that was extracted and adapted from data of (Shah *et al.*, 2012). Three specific behaviors are discriminated in the curve as follows (Baley, 2002; Charlet *et al.*, 2007):

- First linear elastic behavior that corresponds to first mechanical response of the initial cellulose microfibrils structure;
- Non-linear plastic behavior that corresponds to rearrangement of cellulose microfibrils by the tensile motion to be as parallel as possible to the fiber axis;
- Second linear elastic behavior that corresponds to the second mechanical response of the cellulose microfibrils structure after rearrangement.

The alignment of cellulose microfibrils induces rearrangements in the core of the surrounding amorphous polymers and, therefore, the cellulosic reorganization implies an elasto-visco-plastic deformation (Charlet *et al.*, 2007). Consequently, natural fibers cannot be modeled as synthetic fibers (glass and carbon) using an elastic behavior. In the case of flax fibers, a plastic behavior should be considered before fiber failure for the FE model.

Natural fibers show a high variability of their mechanical properties for the same reasons shape a diameter variability discussed in Section “Introduction”. For flax fibers, the literature gives a tensile modulus from 27 GPa to 103 GPa, a tensile strength from 343 MPa to 2000 MPa, and an elongation from 1.2% to 3.3% (Dittenber and GangaRao, 2012). Flax fiber shear strength has been found between 10 MPa and 50 MPa (Panamootil *et al.*, 2017). Therefore, the considered mechanical parameters should be chosen to be as closer as possible to those of flax fibers used in the experimental validation of the model. **Table 1** summarizes the considered mechanical parameters for flax fibers.

The longitudinal plastic behavior of flax fibers is implemented using 30 points on the yield stress versus plastic strain curve from **Fig. 2(b)** with a maximum yield stress of 750 MPa. To model the anisotropic plasticity-based failure for flax fibers, the Hill’s potential function is used (Cardoso and Adetoro, 2017). Hill’s potential function is a simple extension of the Mises function,

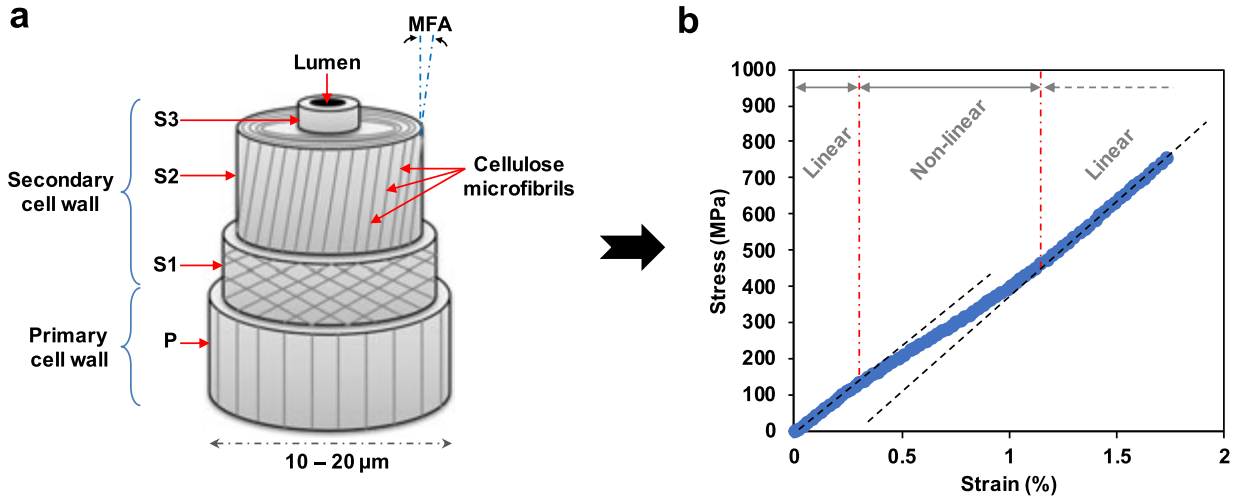


Fig. 2 (a) Schematic illustration of the cellulosic structure of an elementary flax fiber. (b) Typical stress/strain curve of elementary flax fiber.

Table 1 Mechanical properties of elementary flax fibers implemented in the FE model

Property	Direction	Value	Unit	Reference
Stiffness	E_{11}	50	GPa	(Panamoottil <i>et al.</i> , 2017)
	$E_{22} = E_{33}$	12		
	$G_{12} = G_{13} = G_{23}$	3.4		
Poisson ratio	ν_{12}	0.178	–	
	$\nu_{13} = \nu_{23}$	0.2		
Strength	S_{11}	750	MPa	
	$S_{22} = S_{33}$	150		
	$S_{12} = S_{13} = S_{23}$	20		

which can be expressed in terms of rectangular Cartesian stress components (σ_{ij}) as following (Abaqus Analysis User's Manual, 2011; Panamoottil *et al.*, 2017):

$$f(\sigma) = \sqrt{F(\sigma_{22} - \sigma_{33})^2 + G(\sigma_{33} - \sigma_{11})^2 + H(\sigma_{11} - \sigma_{22})^2 + 2L\sigma_{23}^2 + 2M\sigma_{31}^2 + 2N\sigma_{12}^2} \quad (1)$$

Where F , G , H , L , M , and N are constants defined as following:

$$F = \frac{(\sigma^0)^2}{2} \left(\frac{1}{\bar{\sigma}_{22}^2} + \frac{1}{\bar{\sigma}_{33}^2} + \frac{1}{\bar{\sigma}_{11}^2} \right) = \frac{1}{2} \left(\frac{1}{R_{22}^2} + \frac{1}{R_{33}^2} + \frac{1}{R_{11}^2} \right) \quad (2)$$

$$G = \frac{(\sigma^0)^2}{2} \left(\frac{1}{\bar{\sigma}_{33}^2} + \frac{1}{\bar{\sigma}_{11}^2} - \frac{1}{\bar{\sigma}_{22}^2} \right) = \frac{1}{2} \left(\frac{1}{R_{33}^2} + \frac{1}{R_{11}^2} - \frac{1}{R_{22}^2} \right) \quad (3)$$

$$H = \frac{(\sigma^0)^2}{2} \left(\frac{1}{\bar{\sigma}_{11}^2} + \frac{1}{\bar{\sigma}_{22}^2} - \frac{1}{\bar{\sigma}_{33}^2} \right) = \frac{1}{2} \left(\frac{1}{R_{11}^2} + \frac{1}{R_{22}^2} - \frac{1}{R_{33}^2} \right) \quad (4)$$

$$L = \frac{3}{2} \left(\frac{\tau^0}{\bar{\sigma}_{23}} \right)^2 = \frac{3}{2R_{23}^2} \quad (5)$$

$$M = \frac{3}{2} \left(\frac{\tau^0}{\bar{\sigma}_{13}} \right)^2 = \frac{3}{2R_{13}^2} \quad (6)$$

$$N = \frac{3}{2} \left(\frac{\tau^0}{\bar{\sigma}_{12}} \right)^2 = \frac{3}{2R_{12}^2} \quad (7)$$

where each $\bar{\sigma}_{ij}$ is the measured yield stress value when σ_{ij} is applied as the only nonzero stress component, $\tau^0 = \frac{\sigma_0}{\sqrt{3}}$ where σ_0 is the reference yield stress which is set to be equal to σ_{11} in this case; R_{ij} are anisotropic yield stress ratios that are needed to implement

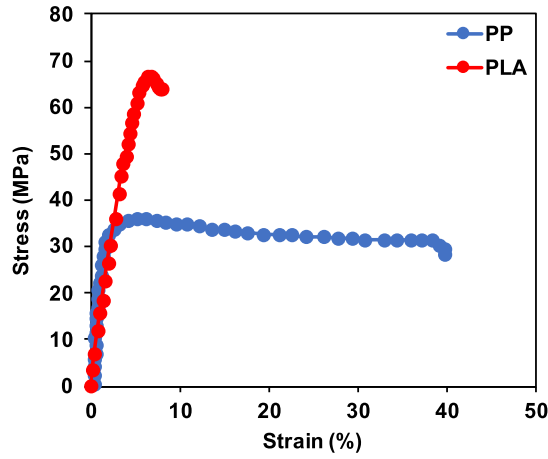


Fig. 3 Typical stress/strain curve of PP and PLA. Adapted from Lin, S., Xia, Y., Lin, C., Wang, J., Gu, G., 2013. Stress state dependent failure loci of a talc-filled polypropylene material under static loading and dynamic loading. In: Proceedings of the 13th International Conference on Fracture, pp. 1–16. Beijing. Yang, J., Pan, H., Sun, S., *et al.*, 2017. A study on the mechanical, thermal properties and crystallization behavior of poly(lactic acid)/thermoplastic poly(propylene carbonate) polyurethane blends. RSC Adv. 7, 46183–46194. doi:10.1039/C7RA07424G.

the Hill's potential function in the FE model (Abaqus Analysis User's Manual, 2011) and are defined as follows where the considered strength values are provided in Table 1:

$$R_{11} = \frac{\bar{\sigma}_{11}}{\sigma^0} \quad (8)$$

$$R_{22} = \frac{\bar{\sigma}_{22}}{\sigma^0} \quad (9)$$

$$R_{33} = \frac{\bar{\sigma}_{33}}{\sigma^0} \quad (10)$$

$$R_{12} = \frac{\bar{\sigma}_{12}}{\tau^0} \quad (11)$$

$$R_{13} = \frac{\bar{\sigma}_{13}}{\tau^0} \quad (12)$$

$$R_{23} = \frac{\bar{\sigma}_{23}}{\tau^0} \quad (13)$$

A ductile criterion (Hooputra *et al.*, 2004) is considered to model the failure of flax fibers. The ductile criterion is based on a fracture diagram which gives the equivalent plastic strain at fracture as a function of the stress state. The model assumes that the equivalent plastic strain at the onset of damage is a function of stress triaxiality and strain rate. The criterion for damage initiation is met when the following condition is satisfied (Hooputra *et al.*, 2004):

$$\int_0^{\varepsilon_{eq}^{**}} \frac{d\varepsilon_{eq}}{\varepsilon_{eq}^{**}(\eta, \dot{\varepsilon}^{pl})} = 1 \quad (14)$$

where ε_{eq} is the equivalent plastic strain, ε_{eq}^{**} is the equivalent plastic strain at fracture, $\dot{\varepsilon}^{pl}$ is the equivalent plastic strain rate, and η is stress triaxiality that can be calculated by the following equations (Danas and Ponte Castañeda, 2012):

$$\eta = \frac{\sigma_m}{\sigma_{eq}} \quad (15)$$

Here σ_m denotes the hydrostatic stress and σ_{eq} is the Von Mises equivalent stress that can be calculated by the following equations:

$$\sigma_m = \frac{\sigma_{11} + \sigma_{22} + \sigma_{33}}{3} \quad (16)$$

$$\sigma_{eq} = \sqrt{\frac{1}{2} [(\sigma_{11} - \sigma_{22})^2 + (\sigma_{22} - \sigma_{33})^2 + (\sigma_{33} - \sigma_{11})^2 + 6(\sigma_{12}^2 + \sigma_{23}^2 + \sigma_{31}^2)]} \quad (17)$$

As for the Hill's potential function, the stress triaxiality criterion is calculated using the strength values of Table 1.

The damage evolution law describes the rate of degradation of the material stiffness once the corresponding initiation criterion has been reached. For elastoplastic materials, the damage evolution is carried out in two forms: softening of the yield stress and degradation of the elasticity (Abaqus Analysis User's Manual, 2011). The damage evolution law can be specified either in terms of

Table 2 Mechanical properties of polymer matrices implemented in the FE model

Material	Property	Value	Unit	Reference
PP matrix	Stiffness	2.1	GPa	(Lin <i>et al.</i> , 2013)
	Poisson ratio	0.4	–	
	Yield stress	35	MPa	
	Strength	30	MPa	
PLA matrix	Stiffness	1.4	GPa	(Yang <i>et al.</i> , 2017)
	Poisson ratio	0.35	–	
	Yield stress	50	MPa	
	Strength	64	MPa	

fracture energy per unit area (G_f) or the equivalent plastic displacement (u^{pl}) which is related to the equivalent plastic strain (ϵ^{pl}) by the following equation (Rao *et al.*, 2007b):

$$u^{pl} = L_e \epsilon^{pl} \quad (18)$$

L_e is a characteristic length related to the element. The definition of the characteristic length depends on the element geometry and formulation: it is a typical length of a line across an element for a first-order element, and it is half of the same typical length for a second-order element (Abaqus Analysis User's Manual, 2011).

The equivalent plastic displacement at failure is computed following the Eq. (19) where σ_{y0} is the value of the yield strength (Abaqus Analysis User's Manual, 2011; Rao *et al.*, 2007b).

$$u_f^{pl} = \frac{2 G_f}{\sigma_{y0}} \quad (19)$$

Finite Element Modeling of Polymer Matrix

The two considered matrices (PP and PLA) are modeled in this study with the same elastoplastic behavior and ductile damage criterion as for flax fibers described in Section "Finite Element Modeling of Natural Fibers". The plastic behavior of the two matrices is also implemented using several points on the yield stress versus plastic strain curve derived from Fig. 3. However, the polymer matrices are assumed to have an isotropic behavior and Hill's potential function is not considered for the plastic modeling. It can be noticed that PP and PLA do not generate the same plastic behavior. PP matrix exhibits a high plasticity that reach 40% of strain before failure, while PLA does not exceed 10%. The mechanical parameters considered in model for the two polymer matrices are given in Table 2.

Finite Element Modeling of Cohesive Interfaces

Interfaces are modeled using the cohesive zone model (CZM). The elastic behavior of the CZM is modeled using a linear elastic traction-separation behavior. It assumes initially linear elastic behavior followed by the initiation and evolution of damage. The elastic behavior is written in terms of an elastic constitutive matrix that relates the nominal stresses to the nominal strains across the interface following the equation (Abaqus Analysis User's Manual, 2011):

$$\begin{Bmatrix} t_n \\ t_s \\ t_t \end{Bmatrix} = \begin{bmatrix} E_{nn} & E_{ns} & E_{nt} \\ E_{ns} & E_{ss} & E_{st} \\ E_{nt} & E_{st} & E_{tt} \end{bmatrix} \begin{Bmatrix} \epsilon_n \\ \epsilon_s \\ \epsilon_t \end{Bmatrix} \quad (20)$$

Where t_n , t_s and t_t are the nominal stresses in the normal, shear and tangential directions, respectively. E_{ij} are the components of the elasticity matrix. ϵ_n , ϵ_s and ϵ_t are the nominal strains in the normal, shear and tangential directions, respectively. The corresponding separations are denoted by δ_n , δ_s and δ_t and are defined as following, where T_0 is the initial thickness of the cohesive element (Abaqus Analysis User's Manual, 2011):

$$\epsilon_n = \frac{\delta_n}{T_0} \quad (21)$$

$$\epsilon_s = \frac{\delta_s}{T_0} \quad (22)$$

$$\epsilon_t = \frac{\delta_t}{T_0} \quad (23)$$

In Abaqus software, the penalty stiffness parameter (K_{ij}) is implemented to model the rigidity of the cohesive zone (Camanho *et al.*, 2003; Park *et al.*, 2016) and it is defined by the following equation:

Table 3 Mechanical properties of cohesive interfaces implemented in the FE model

Interface	Property	Direction	Value	Unit	Reference
Flax/PP	Stiffness	$K_{nn} = K_{ss} = K_{tt}$	27.96	GPa/mm	(Panamoottil <i>et al.</i> , 2017)
		$K_{ns} = K_{nt} = K_{st}$	0		
	Strength	$t_n^0 = t_s^0 = t_t^0$	28.5	MPa	
	Fracture energy	G^C	4	J/m ²	
Flax/PLA	Stiffness	$K_{nn} = K_{ss} = K_{tt}$	38	GPa/mm	Calculated from data of (Le Duigou <i>et al.</i> , 2014)
		$K_{ns} = K_{nt} = K_{st}$	0		
	Strength	$t_n^0 = t_s^0 = t_t^0$	18	MPa	
	Fracture energy	G^C	28	J/m ²	(Le Duigou <i>et al.</i> , 2014, 2010)

$$\begin{Bmatrix} t_n \\ t_s \\ t_t \end{Bmatrix} = \begin{bmatrix} K_{nn} & K_{ns} & K_{nt} \\ K_{ns} & K_{ss} & K_{st} \\ K_{nt} & K_{st} & K_{tt} \end{bmatrix} \begin{Bmatrix} \delta_n \\ \delta_s \\ \delta_t \end{Bmatrix} \quad (24)$$

Damage of cohesive zones is assumed to initiate when a quadratic interaction function involving the nominal stress ratios reaches a value of one. This criterion can be represented as following ("Abaqus Analysis User's Manual," 2011; Panamoottil *et al.*, 2017):

$$\left(\frac{\langle t_n \rangle}{t_n^0} \right)^2 + \left(\frac{t_s}{t_s^0} \right)^2 + \left(\frac{t_t}{t_t^0} \right)^2 = 1 \quad (25)$$

t_n^0 , t_s^0 and t_t^0 are the peak values of the nominal stress components in the normal, shear and tangential directions, respectively. The symbol $\langle \rangle$ in the Eq. (25) represents the Macaulay bracket which is used to signify that a pure compressive deformation or stress state does not initiate damage ("Abaqus Analysis User's Manual," 2011). Table 3 gives the considered mechanical parameters of the cohesive element for both flax/PP and flax/PLA interfaces.

Experimental Validation

The experimental validation of the FE model has been performed using orthogonal cutting process on both flax/PP and flax/PLA composites. The two NFC samples are manufactured by thermocompression of unidirectional flax fabrics and polymer matrix. Flax fabrics are based on technical fibers that are a bundle of about 3–6 elementary fibers.

Orthogonal cutting tests are performed on a shaper machine (GSP – EL 136) with a carbide cutting insert (Sandvik – TCGX 16 T3 04 – AL H10) as shown in Fig. 4. The machining forces are acquired using a piezoelectric dynamometer (Kistler – 9255B). The machined surfaces are then observed at microscale using the scanning electron microscope (SEM) at low vacuum mode (JEOL – 5510LV). To ensure the repeatability of the experimental outputs, each cutting configuration is tested three times. Thus, the final outputs from the orthogonal cutting experiments are presented as the mean value of these three repeated tests. Measurement errors are considered as the average of the absolute deviations of data repeatability tests from their mean.

The cutting experiments on flax/PP samples have been devoted to investigating the effect of cutting speed (V_c). Therefore, the cutting speed has been varied from 12 m/min to 80 m/min. On the other side, the cutting experiments on flax/PLA samples concerned the study of the fiber orientation effect (θ). To this aim, the orientation of flax fibers, with respect to the cutting direction, has been varied from 0° to 90°. Table 4 summarizes the experimental cutting conditions related to the tool geometry, tool kinematic, and the process parameters. The FE model was performed with the same cutting conditions.

Numerical Effect of Cutting Speed

The numerical investigation of the effect of cutting speed has been performed on flax/PP composites with a fiber orientation of 90° from the cutting direction (Chegdani *et al.*, 2019b). Fig. 5 presents the comparison between the numerical and the experimental machining results of flax/PP at low cutting speed (12 m/min) and high cutting speed (80 m/min). It can be noticed a significant impact of the cutting speed on the cutting behavior of flax fibers inside the composite. Cutting with low cutting speed induced high transverse plastic deformation of flax fibers toward the cutting direction. Flax fibers are then torn off rather than sheared as shown in Fig. 5(a) and (c). Consequently, high decohesion zones are produced because of interfaces damage. When increasing the cutting speed to high values, flax fibers are sheared efficiently thanks to the increase of the contact stiffness. Flax fibers are then less deformed and the decohesion zones are strongly reduced as shown in Fig. 5(b) and (d). All these cutting phenomena related the cutting speed effect are reproduced by the FE model that shows also a less induced subsurface damage when cutting with high cutting speed.

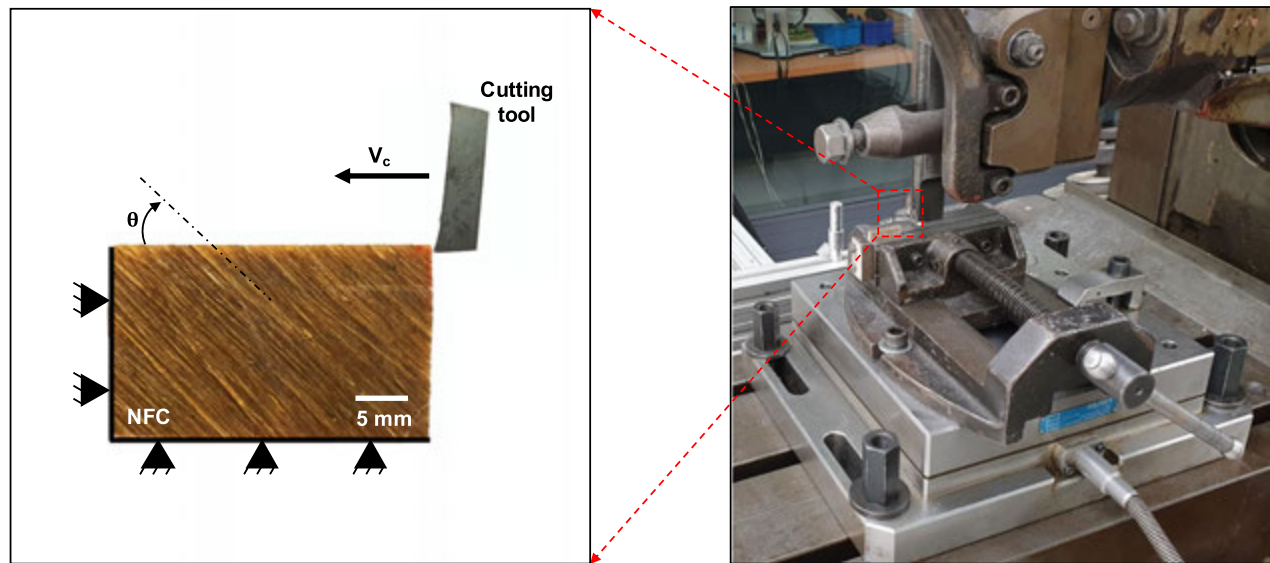


Fig. 4 Photographic image of the experimental setup with a zoom on the active zone of tool/material interaction.

Table 4 Experimental cutting condition for orthogonal cutting tests

		Parameter	Value	Unit
Tool geometry		Rake angle (γ)	20	Degree ($^{\circ}$)
		Clearance angle (α)	7	Degree ($^{\circ}$)
		Edge radius (r_e)	12	μm
Cutting parameters	Flax/PP cutting	Depth of cut (a_p)	100	μm
		Fiber orientation	90	$^{\circ}$
		Cutting speed (V_c)	12/20/32/50/80	m/min
	Flax/PLA cutting	Depth of cut (a_p)	100	μm
		Fiber orientation	0/25/45/65/90	$^{\circ}$
		Cutting speed (V_c)	50	m/min

Fig. 6 presents a comparison between the experimental and the numerical machining forces obtained for the orthogonal cutting of flax/PP composites. The numerical cutting forces correspond to the experimental ones in terms of trend and magnitude where the cutting forces increase by increasing the cutting speed (**Fig. 6(a)**). For the thrust forces, the numerical outputs correspond to the experimental ones in terms of trend. However, there is a factor of 3 between the experimental and the numerical values as clearly shown in **Fig. 6(b)**. This correlation issue is well known also in FE machining models of synthetic fiber composites (Arola and Ramulu, 1997; Lasri *et al.*, 2009) and it can be due to the fibers spring-back as reported in (Lasri *et al.*, 2009). The magnitude factor between simulation and experimental thrust forces is around 8.5 in (Arola and Ramulu, 1997) and around 9.33 in (Lasri *et al.*, 2009) for the same considered fiber orientation (90°). The magnitude factor of 3 found in the current model for machining NFC is less than those of the synthetic fiber composite model because natural fibers have less spring-back intensity due to their high transverse elasticity (Chegdani *et al.*, 2019b). Therefore, next section will address this correlation issue through a tribological approach based on the friction properties of the tool/NFC cutting contact.

Numerical Effect of the Local Cutting Friction

In Section “Numerical Effect of Cutting Speed”, the FE model shows correlation issues of the thrust forces that can be due to the spring-back phenomenon of fibers during the cutting operation. However, no theoretical or experimental evidence can confirm this hypothesis. For this reason, a different approach is investigated in this section to address the issue of the thrust force correlation in the FE modeling of NFRP machining. This approach is based on the machining tribology, especially the micro-friction between the composite and the cutting tool. The micro-friction means the local friction between the cutting tool and each composite phase component at microscale (elementary natural fibers and polymer matrix). The considered investigation is

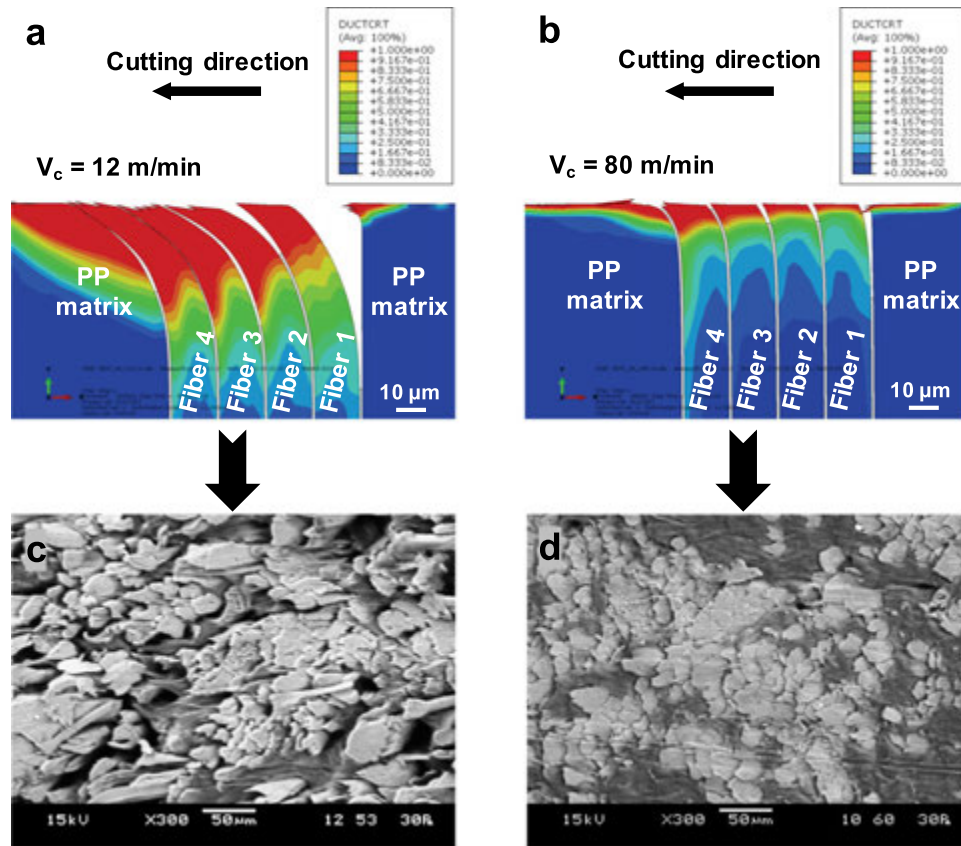


Fig. 5 Numerical and experimental cutting behavior of flax fibers within the flax/PP composite at low and high cutting speed.

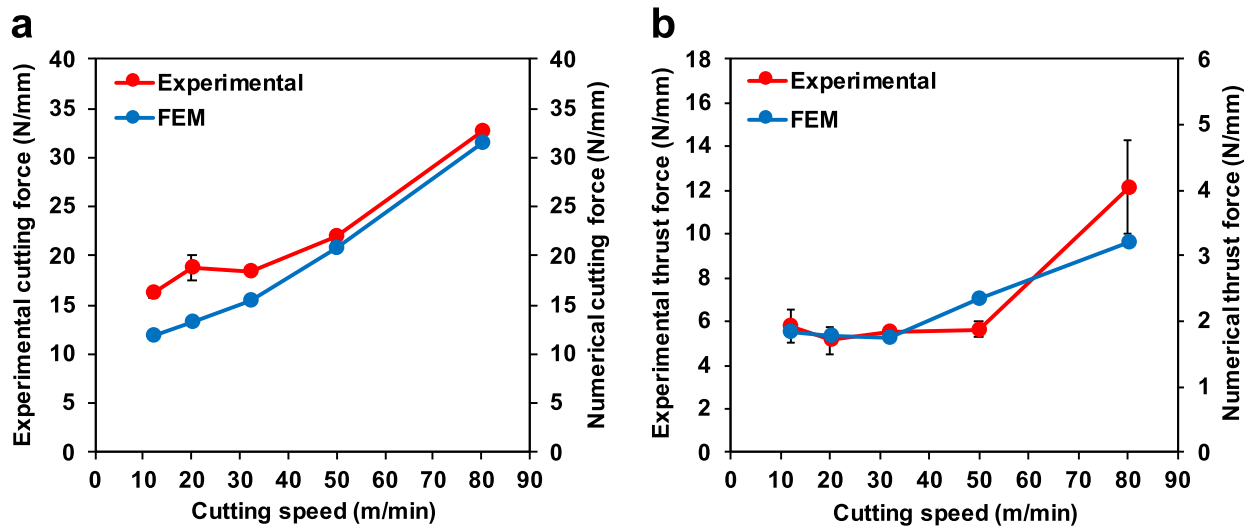


Fig. 6 Comparison between numerical and experimental machining forces for flax/PP composites. (a) cutting forces, (b) thrust forces.

motivated by previous works on the local micro-friction on NFC composites by scratch-test experiments (Chegdani *et al.*, 2019a, 2018b). Indeed, the first scratch-test experiments were done with a diamond Berkovich tip indenter and the friction coefficient has been found around 0.4 for polypropylene (PP) matrix and around 0.5 for flax fibers (Chegdani *et al.*, 2018b). These values were used in the previous FE model of machining (Chegdani *et al.*, 2019b). However, the following scratch-test experiments were done with a Sapphire Berkovich tip indenter and the friction coefficient has been found around 0.2 for both PP matrix and flax fibers at the same scratching conditions as the experiments with the diamond indenter (Chegdani *et al.*, 2019a). This indicates that the tool material causes a significant modification of the frictional properties in the case of NFRP composites. Therefore, the values of the

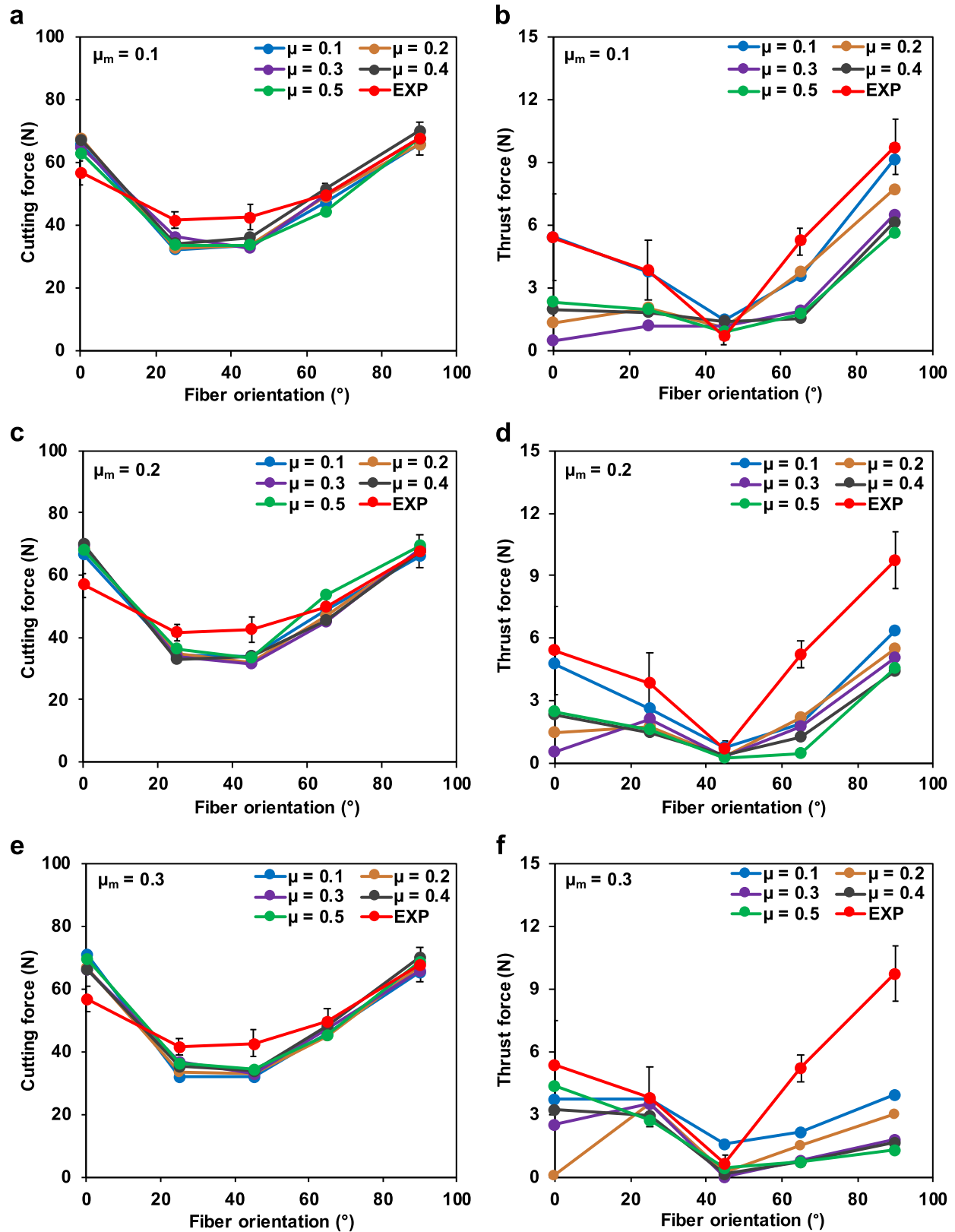


Fig. 7 Comparison between experimental and numerical machining forces at different values of micro-friction. (a, c, e) cutting forces and (b, d, f) thrust forces. μ in the graphs corresponds to μ_f .

micro-friction used in the previous FE machining model may be not appropriate because the experimental machining tests were performed by a carbide cutting insert. This could be the origin of the magnitude difference in the thrust forces of the FE machining model.

To this aim, the FE micromechanical model is used in this section for flax/PLA composites to investigate the effect of the local micro-friction on the cutting behavior for different fiber orientation values.

To simulate the orthogonal cutting friction with ABAQUS/CAE software in the micromechanical FE model, a penalty contact method is considered to define each contact interaction (*Abaqus Analysis User's Manual, 2011*). This method is based on Coulomb friction law to control the frictional contacts between the cutting tool and the composite. Coulomb friction law assumes that relative motion between the tool and the composite occurs at the contact point when the equivalent shear stress along the tool-material interface (τ_f) is more than or equal to the critical friction stress as follows:

$$\tau_f \geq \tau_{crit} = \mu n_p \quad (26)$$

Where μ is the friction coefficient and n_p is the normal pressure at the same point.

The notion of friction in the FE model is based on the interaction between the nodes of the two surfaces in contact. Since the developed model is performed at microscale, the friction coefficient needed for the micromechanical model should be the local micro-friction coefficient at each point of the cutting contact. Thus, the micromechanical model will differentiate two contact pairs with the cutting tool because elementary flax fibers and PLA matrix are modeled separately. Consequently, two micro-friction parameters will be considered in this study:

- The micro-friction coefficient of the contact between the cutting tool and flax fibers (μ_f)
- The micro-friction coefficient of the contact between the cutting tool and PLA matrix (μ_m)

The tested values of micro-friction coefficients has been chosen to vary from 0.1 to 0.5 based on the results of the scratch-test experiments (*Chegdani et al., 2019a, 2017*).

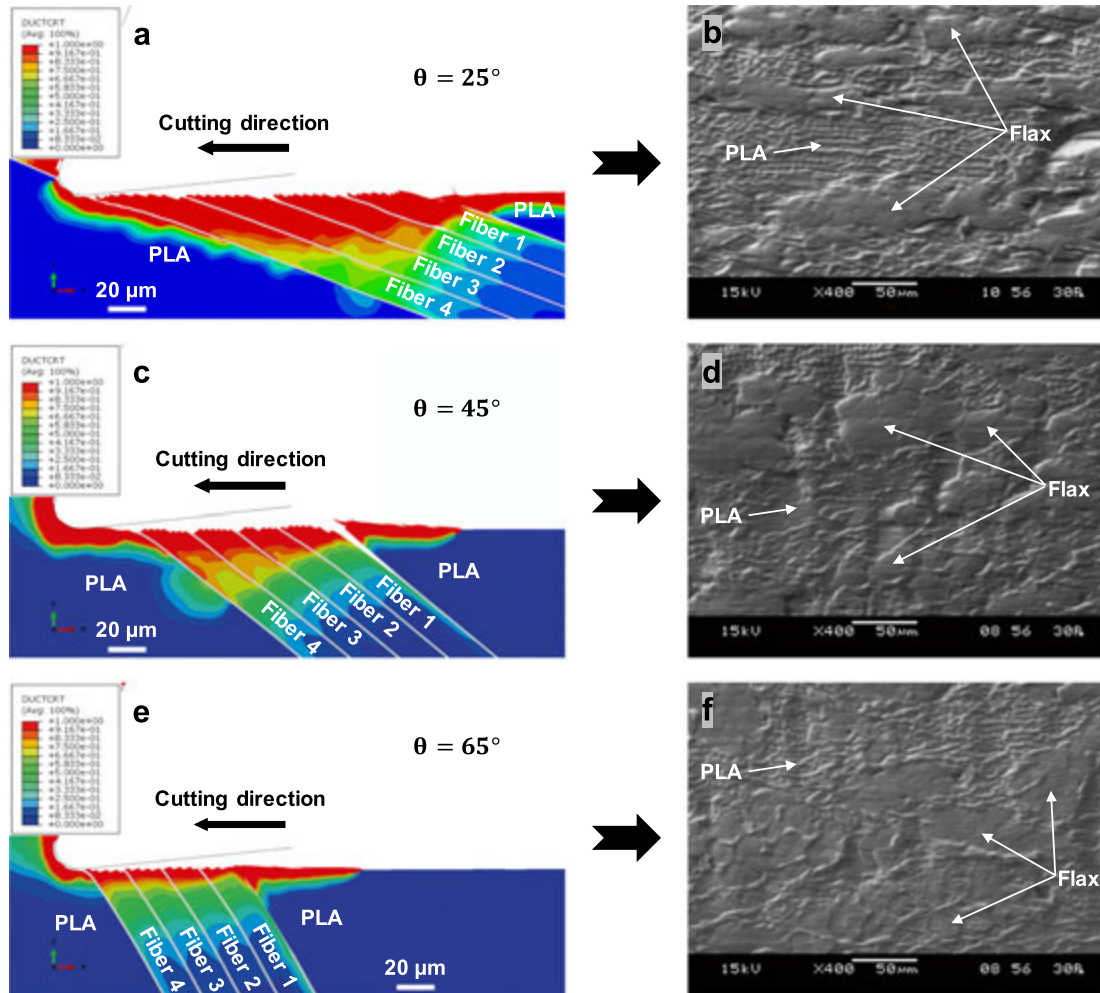


Fig. 8 Numerical and experimental cutting behavior of flax fibers within the flax/PLA composite at different values of fiber orientation.

Fig. 7 shows the comparison between numerical and experimental machining forces with different values of micro-friction coefficients. In each graph of **Fig. 7**, the micro-friction coefficient of the PLA matrix (μ_m) is kept constant, and the micro-friction coefficient of flax fibers (μ_f) is varied from 0.1 to 0.5 in order to evaluate its effect on the numerical machining forces.

Generally, the cutting forces are not significantly affected by the variation of the numerical micro-friction coefficients. As for the numerical results of **Fig. 6**, the numerical cutting forces correspond to the experimental outputs in terms of trend and magnitude as shown in **Fig. 7(a)**, **(c)**, and **(e)**.

On the other side, the numerical thrust force outputs are highly dependent on the variation of the micro-friction coefficients in the FE model. Indeed, when performing the FE simulations with $\mu_m = 0.1$, it can be seen in **Fig. 7(b)** that numerical thrust forces are closer to the experimental ones at the lowest value of micro-friction coefficient of flax fibers with the cutting tool ($\mu_f = 0.1$). Increasing μ_f provokes a deviation of the numerical thrust forces from the experimental outputs.

Furthermore, Increasing the value of μ_m leads to rise the divergence between the numerical and the experimental values of thrust forces (**Fig. 7(b) → d → f**). Therefore, **Fig. 7** indicates that the FE model can reproduce the experimental outputs with good accuracy for an isotropic micro-friction where $\mu_f = \mu_m = 0.1$.

Numerical Effect of Fiber Orientation

In this section, the numerical simulations are performed with an isotropic micro-friction coefficient of 0.1 as suggested by the tribological investigation of Section "Numerical Effect of the Local Cutting Friction". **Fig. 8** presents a comparison between the numerical and the experimental cutting behavior of flax fibers inside the PLA matrix at different values of fiber orientation. Cutting with $\theta = 25^\circ$ induces the highest ductile damages caused by plastic deformation as shown in **Fig. 8(a)**. These damages weaken the fiber shearing which is clearly obvious in SEM image of **Fig. 8(b)** where flax fibers are so deformed that it is not possible to distinguish the elementary fibers in each fibers bundle.

When changing the fiber orientation from 25° to 45° , ductile damages are reduced which enhance the shearing of flax fibers as shown in **Fig. 8(c)**. The same finding is noticed in the corresponding SEM image of **Fig. 8(d)** when elementary flax fibers becomes slightly visible due to the decrease of plastic deformation when cutting.

At $\theta = 65^\circ$, **Fig. 8(e)** the ductile damages are significantly reduced and the fiber shearing are strengthened which corresponds to the experimental observation of **Fig. 8(f)** where no significant plastic deformation of flax fibers is noticed and the cross-sections of elementary fibers are clearly visible.

Conclusions

In this article, a 2D finite element model is developed to simulate the machining behavior of natural fiber composites at the microscopic scale of natural fibers. Flax fiber reinforced polypropylene and flax fiber reinforced polylactic-acid are considered to perform the experimental validation using the fundamental process of orthogonal cutting. Cutting speed and fiber orientation parameters are used to investigate the predictiveness accuracy of the FE model. The following conclusions can be drawn:

- Unlike synthetic fibers such as glass and carbon, natural fibers should be modeled with an elasto-plastic behavior and a ductile criterion for failure.
- The FE model is able to predict with good accuracy the qualitative cutting behavior of flax fibers inside the polymer matrix.
- The FE model can predict with good accuracy the cutting forces. However, the thrust forces are predicted with a deviation in the magnitude.
- The magnitude predictiveness issue of thrust forces is optimized by investigations the local micro-friction between the cutting tool on each composite component at microscale. It has been found that an isotropic micro-friction coefficient of 0.1 provides a good predictiveness of the thrust forces.

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Eco-Friendly Composites for Brake Pads From Agro Waste: A Review

Bushra Rashid, Universiti Putra Malaysia, Serdang, Selangor, Malaysia and Middle Technical University, Alzafarany, Baghdad, Iraq
Zulkiflle Leman, Mohammad Jawaid, and Mohamad R Ishak, Universiti Putra Malaysia, Serdang, Selangor, Malaysia
Faris M Al-Oqla, The Hashemite University, Zarqa, Jordan

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Introduction

Eco-Friendly Solution for Industry

Proper waste management and utilization have recently become essential for both industrial sustainability and the environment. Large quantities of agro waste fibers are accumulated annually without benefits, and some of them are burned (Rashid *et al.*, 2017b; Al-Oqla *et al.*, 2015e). Besides, coal fired power plants around the world produce very large amounts of fly ash every year, 70 million tons of which are generated in the United States alone. Only 40% of all fly ash produced in the United States find useful applications and the rest must be disposed of; this is burned for the generation industry and is dramatically involved in environmental pollution as well as ecological problems (Malhotra *et al.*, 2002). Moreover, utilizing various agro waste types in useful applications is not widely used. Beside the recent international effort on environmental concerns and the need to create a better environment, the usage of natural fibers has attracted attention for many applications (Shalwan and Yousif, 2013; Njuguna *et al.*, 2011; Koronis *et al.*, 2013). In addition to the environmental benefits, cellulose-based materials can also improve physical, mechanical, and tribological properties, while reducing costs of raw material and enhancing the cleaner production as well as the industrial sustainability (Rashid *et al.*, 2017a; Al-Oqla *et al.*, 2015a). The current trend in the research field is to the utilization of industrial or agricultural wastes as a source of raw materials for composite development. The attractive performance-to-cost ratio involved in producing composite materials from wastes to create brake pads has inspired the idea of discovering the possible combination of additional waste materials in these preparations (Ruzaidi *et al.*, 2011b). This will provide more economical benefit and also environmental preservation by utilizing the waste of natural fiber.

Natural fibers have been used to reinforce materials for more than 1000 years (Menezes *et al.*, 2012). Also, composites have developed as a cost-effective and potentially environmentally friendly alternative to synthetic fiber reinforced composites. Moreover, instead of utilizing synthetic fibers, or their combinations with different types of natural ones, engineers have used the natural fibers to meet the required design for different applications because of their potential to replace the traditional synthetic fibers (Al-Oqla *et al.*, 2015d,c; Suddell, 2008; Aridi *et al.*, 2016). Nowadays, the markets for natural fiber are growing and their number of uses are increasing, as it is time to change toward sustaining the environment and find renewal and nontoxic resources for industry (Njuguna *et al.*, 2009). Meanwhile, brake pads are one of the most important key factors in the competition to develop greener transport (Nosonovsky and Bhushan, 2012). The availability of natural fibers as well as their comfort in the industrial usage have attracted researchers to study their possibility of reinforcement and suitability to fulfill the essential specifications for better reinforced polymer composite for tribological as well as other applications (Al-Oqla *et al.*, 2015b,e). Many studies have been made using natural fibers such as flax, bamboo, sisal, hemp, banana, and sugarcane fibers as reinforcements in brake pad applications (Sloan *et al.*, 2006; El-Tayeb, 2008; Aigbodion *et al.*, 2010; Fu *et al.*, 2012; Idris *et al.*, 2015; Lee and Filip, 2013; Maleque and Atiqah, 2013).

In the automotive industry, free asbestos, Cu, and Sb fibers and other toxic ingredient-based components like lining couplings and brake pads, etc. have been needed because of human health and environmental concerns. Other alternatives to these fibers include metal fiber, mineral fiber, and artificial polymer fibers such as alumina fiber, glass fiber, steel fiber, carbon fiber, aramid fiber, and their combinations. Recently, wide ranging research has concentrated on the possible use of natural fiber agricultural products as alternative fibers in eco-friendly brake pad composites (Chin and Yousif, 2009a; Mutlu, 2009b; Menezes *et al.*, 2012; Bajpai *et al.*, 2013). Hence, this article presents a comprehensive review of these studies and suggests a direction for future developments.

Development of Brake Pad Composites

The brake system is the most important safety part of a vehicle; it must stop the automobile quickly and dependably under different conditions. Brake pads, as shown in Fig. 1, mainly change the kinetic energy of the vehicle to thermal energy by friction (Verma *et al.*, 2015). When the brakes are hydraulically functioned, the caliper squeezes or clamps the two pads together into the rotating rotor to slow/stop the vehicle. The brake caliper contains two brake pads with their friction surfaces facing the rotor. As the pad is heated due to the rotor contact friction, small amounts of friction material are transferred to the disk, converting it to a dull gray. As a result, the disk and brake pad (both now with friction material), then “stick” to each other, offering the friction that stops the vehicle (Fu *et al.*, 2012).

Braking is accompanied with the release of a variety of particulates. Automotive brake pads available in the market are characterized as metallic, semimetallic, or nonasbestos organic (NAO) materials (Blau, 2001; Chan and Stachowiak, 2004; Fu *et al.*, 2012). They are considered as organic friction materials because the matrix of these composites consists of one or more



Fig. 1 Brake pads (Brake pad, 2014).

polymers. Automotive brake materials are complex composites containing a mixture of many ingredients; more than 2000 different elements (typically 5–30) are required for the optimization of the friction and wear behavior (Dahlke *et al.*, 1998; Vaculik *et al.*, 2014; Roubicek *et al.*, 2008; Neis *et al.*, 2014).

The first group of recent brake friction composites was asbestos reinforced composites and it has been banned because of human health and harmful environmental issues. Asbestos was recognized as harmful content as it causes heart and lung cancer, as well as other health problems. Since the 1970s, asbestos has been commonly acknowledged as a carcinogen, though the introduction of the asbestos ban in the United States only came about in 1989 (Menezes *et al.*, 2012). Consequently, due to the environmental problems connected to its fibers, the asbestos, which contains friction composites, was changed to NAO and semimetallic composites. Though use of asbestos for brake pads has been forbidden (Menezes *et al.*, 2012; Deepika *et al.*, 2013; Shalwan and Yousif, 2014), many in the brake pad industry are getting away from asbestos brake pads due to concerns regarding airborne particles in the factories and discarding of wastes that contain asbestos. Moreover, according to the rules against hazardous ingredients in the United States and Europe, numerous raw materials frequently used in commercial friction materials might have a possible negative environmental impact. Furthermore, components like copper, antimony trisulfide, lead, potassium titanate whisker, tin, and others that have a negative environmental impact are regularly discussed.

Besides the elimination of toxic ingredients, the eco-friendly brake materials are developed using geopolymer matrix and natural fiber waste to replace phenolic resin and synthetic fibers, respectively (Mathur *et al.*, 2004; Surojo *et al.*, 2014). Both the binder resin and reinforcing fibers can significantly determine the friction characteristics of the produced material (Kim *et al.*, 2008; Bijwe, 2007). Friction materials were commonly established over trial and error, combined with earlier experience of producers (Lu *et al.*, 2010). Lately, a few mathematical methods have been proposed for optimization and evaluation of novel brake composites. Yet, they are not being widely used by producers, and still remain as academic tools (Yun *et al.*, 2010; Zaharudin *et al.*, 2012).

Selection of Ingredients for Brake Pad

Several features and criteria have to be considered in selecting the brake pad material. The most important are the following:

1. The capability of the material to resist brake fade at higher temperatures (Lee and Filip, 2013; Matějka *et al.*, 2013).
2. The effects of water on brake fade (all brakes are designed to stand at least temporary contact to water) (El-Tayeb and Liew, 2009).
3. The material must show suitable values of friction factor and also better resistance to wear (Mutlu, 2009b; Matějka *et al.*, 2013).
4. The thermal stability and the ability to recover rapidly from either high temperature or moisture (Lee, 2013).
5. The ability of the material to provide smooth, even contact with the rotor or drum (rather than a material that breaks off in chunks or causes pits or dents) (Owen, 2011).

Most of the brake composites are usually comprised of different materials. These ingredients frequently comprise many disparate ingredients such as polymers, ceramics, and metals to satisfy several essential requirements such as wear resistance and high temperature friction stability under several functioning parameters such as applied loads, speeds, and temperature. In addition, it should have no vibration and no noise at wide ranges of braking conditions, as well as low cost (Blau, 2001; Bhane *et al.*, 2014; Talib *et al.*, 2012). Also, the friction materials are required to provide environmental conditions (Lee, 2013). Nevertheless, it is almost impossible to have all these preferred features. Thus, some necessities have to be compromised to reach some other requirements. Generally, each design of friction material has its own unique wear-resistance characteristics and frictional behaviors. Moreover, the hybridization technique has been acknowledged as able to provide the balance between performance, cost, and more recently environmental attributes for natural fiber composites in many specific applications (Mansor *et al.*, 2014; Jawaidd and Abdul Khalil, 2011).



Fig. 2 Types of reinforcement fibers.

Nowadays, brake pad materials are categorized as belonging to one of four principal classifications (Chan and Stachowiak, 2004; Blau, 2001; Menezes *et al.*, 2012) as follows:

1. Binder: a type of thermos-resin used to hold all other components together in order to form a thermally stable matrix.
2. Structural materials: typically fibers of metal, carbon, glass, Kevlar, and/or ceramic fibers are utilized to prove mechanical strength. **Fig. 2** shows some types of fibers used in brake pad composites.
3. Fillers: fillers such as mica, vermiculite, and barium sulfate are used mainly to reduce cost of pads.
4. Frictional additives: solid lubricants like graphite and metal sulfides are utilized to ensure stable frictional properties and control wear primarily at elevated temperatures.

There is a little uncertainty or fuzziness in this classification. Some of the additives can be positioned into more than one category since they fulfill numerous functions. Moreover, many factors should be considered in the development of brake materials so as to fulfill the requirements such as lower wear rate and stable friction coefficient at several operating speeds, temperatures, pressures, and environmental conditions in the automotive sectors. This is important to having an appropriate combination of materials in order to have those requirements with reasonable cost of materials. Selection of the material is not an easy task; rather it is a complex process (Adebisi *et al.*, 2011; Maleque and Sarker, 2010; Al-Oqla *et al.*, 2014b, 2016; Rashid *et al.*, 2017c). Accordingly, there are inescapable intersections in the tabular scheduling (Blau, 2001; Liew and Nirmal, 2013). The type and amount of these ingredients are determined mostly based on experience, empirical observation, or a trial and error method to make a new formulation. **Table 1** shows the most ingredients used to fabricate brake pad composites (Chan and Stachowiak, 2004; Han *et al.*, 2006; Hee and Filip, 2005; Lee and Filip, 2013; Yun *et al.*, 2010).

Environmental Issues Related to Toxic Ingredients in Brake Pads

Braking processes generate brake wear debris due to the wear of friction materials and their counterparts (typically rotors). The wear debris from some brake friction materials are proving to be geotaxis and harmful to the environment (Kukutschová *et al.*, 2009). Asbestos fiber used to be the important friction fiber enhanced polymeric composites in brake pads, brake linings, brake couplings, etc. Asbestos is hydrated magnesium silicate $\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4$. Once it is used, the asbestos in the automobile brakes varies from 30 to 70% (Blau, 2001). According to Nicholson, asbestos has significant properties such as thermal stability above 500°C , processes well in terms of friction and wear, and is available and cheap (Nicholson, 1995; Menezes *et al.*, 2012). In 1986, the Environmental Protection Agency proposed a ban on asbestos that required all new vehicles to have nonasbestos brakes by September 1993, and the aftermarket would have had until 1996 to convert to nonasbestos. This is due to evidence by a medical research that asbestos fibers would lodge in the lungs causing adverse respiratory conditions (Blau, 2001). Although the EPA's proposed ban was overturned in federal court, a major shift away from asbestos was considered by most friction material suppliers and vehicle manufacturers.

Although many people believe that asbestos was replaced by NAOs years ago, asbestos brake products are still used commercially. A report showed that 9.5% of the vehicles "serviced by its readers" have asbestos linings installed on them (Blau, 2001). This is an important percentage of the total brake market, since many people believe that asbestos is no longer used. This is because it is a cheap fiber for low temperature brake pad applications, but it has now been voided because of its cancer causing nature. Hence, new asbestos-free brake pads and friction materials have been established (Menezes *et al.*, 2012).

In addition, copper was used in brake materials for decades. Yet, the environmental effect of copper in brake materials was only observed lately. The Brake Pad Partnership (BPP), which is a group of specialists from environmental agencies and brake industries, developed an advanced model for how commercial brake wear wreckage released into the air or onto the streets ends up in the waterways (Hagino *et al.*, 2015). As copper in the brake pad wreckage can be toxic to aquatic organisms, Washington State chartered Senate Bill 6557 to enhance brake friction materials limitations in June 2010 to limit the concentration and the usage of hazardous elements and their compounds in brake friction material (Lee and Filip, 2013). Also, the State of California approved

Table 1 Ingredients of brake pad composites

<i>Binder</i>	<i>Abrasive and lubricant</i>	<i>Reinforcement fiber</i>	<i>Additive and filler</i>
Phenolic resin	Nitrile butadiene rubber (NBR)	Steel wool	Barium sulfate
Geopolymer	Styrene butadiene rubber (SBR)	Potassium titanate whisker (a type of ceramic)	Calcium carbonate (CaCO ₃)
Condensed polynuclear aromatic (COPNA) resin	Coke-carbon	Lapinus fiber	Vermiculite
Silicone-modified phenolic resin	Synthetic graphite	Twaron fiber	Cordierite
Cyanate ester resin	Potassium titanate	Aramid pulp	Mica
Epoxy-modified phenolic	Iron powder (Fe)	Ceramic fiber	Potassium
Thermoplastic polyimide resin	Pyrite (FeS ₂)	Polyacrylonitrile (PAN)	Calcium sulfate
Modified resins	Dolomite	Copper, fiber, copper chips	Cashew dust
Cashew net shell liquid (CNSL)	Clay minerals with nano-titanium dioxide (TiO ₂)	Brass fiber	Copper powder
	Hexagonal boron nitride (H-BN)	Glass	Cashew dust shell liquid
	Metal sulfides (antimony, tin, copper, lead, and sulfides)	Sepiolite	Alkali metal titanates
	Metal oxides/silicates, for example, quartz (SiO ₂), zirconium silicate (ZrSiO ₄), zirconium oxide (ZnO), aluminum oxide, MgO, BaSO ₄ , (Al ₂ O ₃), Sb ₂ S ₃ , etc.	Mineral fiber	Molybdenum trioxide
	Friction dust	Natural fiber	Rubber dust
			Wollastonite
			Talc
			Fly ash

the Senate Bill 346, which forbids motor vehicle brake materials that contain more than 5 and 0.5 wt% copper by January 1, 2021 and January 1, 2025, respectively. Moreover, in 2012, the National Contaminant Biomonitoring Program in the United States Fish and Wildlife Service found that zinc, copper, and lead in the Manoa Stream in Hawaii watershed were anthropogenically generated and the primary contributors were automotive emissions plus vehicle wear.

Nowadays, the growing interest in adopting natural fibers as reinforcement for polymeric composites for industrial applications is increased due to their useful and eco-friendly properties such as nontoxicity, low cost, biodegradability, light weight, renewability, high specific strength, nonabrasivity, and combustibility (Al-Oqla *et al.*, 2014a,b, 2015a). In addition to that, such fibers have high specific properties such as stiffness, impact resistance, flexibility, and modulus. Other properties include less skin and respiratory irritation, vibration damping, excellent sonic insulation properties, and enhanced energy recovery (Chin and Yousif, 2009b; Al-Oqla *et al.*, 2015b; Al-Oqla and Sapuan, 2015). On the other hand, agro waste products are emerging as new and inexpensive materials in friction materials development with commercially viable and environmental acceptability (Mutlu, 2009a).

Novel Green Brake Pad Composites

Many efforts have been carried out to fit the above points by developing new brake pads using different waste materials. Due to the current regulations, the study of environmentally friendly friction materials for brakes is essential. The objectives are to reduce the use of possibly hazardous elements in the formulations of brake material while keeping the friction performance and decreasing wear. Reducing wear of material would reduce the possible impact to the environment (Lee and Filip, 2013). In recent years, many materials are used as an alternative to the toxic ingredients. Due to their good tribological priorities, there are three groups of alternative materials used or that are candidates to use in eco-friendly applications, which are natural fiber, plant powders, and biomaterial powders, as shown in Table 2. In addition, researchers have tried to find new modified resins to use in eco-friendly brake pads (Balaji and Kalaichelvan, 2012; Mathur *et al.*, 2004).

Palm Kernel Fibers and Shell

Ibhadode and Dagwa (2008) developed asbestos-free friction lining material from palm kernel shell (PKS) and phenolic resin. Taguchi optimization technique was used to achieve optimal friction material formulation and manufacturing parameters. The derived friction material was used to produce automobile disk brake pads. The laboratory brake pads were tested for wear and effectiveness on a car. Their performance was considered satisfactory after being compared with a premium asbestos-based commercial brake pad. However, more pad wear was observed on the PKS pad at high vehicular speeds beyond 80 km/h as shown in Fig. 3.

Also, different weight fractions of palm kernel fibers (PKFs) with size of 100 µm were added in varying percentages to other ingredients (calcium carbonate, aluminum oxide, and epoxy polymer) to fabricate the composite by Ikpambese *et al.* (2014). The

Table 2 Alternative materials in eco-friendly brake pad

Natural fiber waste	Shell waste	Biomaterial waste
Flax, JMM, hemp, sugarcane (bagasse), palm kernel, jute, coir, kenaf, sisal, bamboo, cotton, palm, wheat, Nile roses, palm slag, battle net, straw and sunflower, banana peels (powder), rice straw dust, rice husk dust, and <i>Jatropha</i> seed cake	Hazelnut shells, coconut shell	Periwinkle shell, wool, fish bone, chicken feather, and eggshell particles

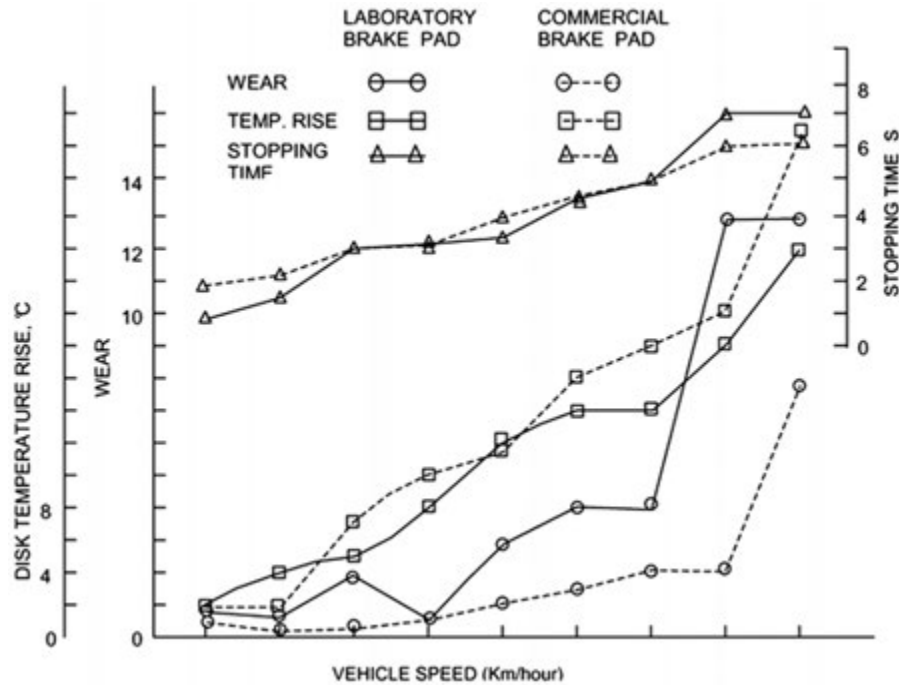


Fig. 3 Comparison of laboratory and commercial brake pads under dynamic testing. Reproduced from Ibhadoke, A.O.A., Dagwa, I.M., 2008. Development of asbestos-free friction lining material from palm kernel shell. *Journal of the Brazilian Society of Mechanical Sciences and Engineering* 30, 166–173.

composites were labeled as S1, S2, S3, S4, S5, and S6. The result shows that wear rate (see Fig. 4), coefficient of friction (see Fig. 5), noise level, temperature, and stopping time of the produced brake pads increased as the speed increased. Moreover, the sample with a composition of 40% epoxy resin, 10% palm wastes, 6% Al_2O_3 , 29% graphite, and 15% calcium carbonate gave better properties. In addition, the values of the necessary parameters obtained from PKFs are within (and even better than) the standard requirement for commercial brake performance pad.

Both studies reported that an increase in speed leads to increase in contact pressure between the rotor and brake pads, thus increasing the wear rate. Table 3 shows the properties of eco-brake pads using PKS and PKF. It was observed that although the wear is higher for eco-brake pads using PKF the coefficient of friction is less compared with eco-brake pads using PKS. Also, stopping time for the composite using PKS is less than that using PKF. These reports confirmed the earlier result obtained by Koya and Fono (2009).

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A new brake pad composite consisting of PKS (which is an agro waste), cashew nut shell liquid, sulfur, calcium carbonate, quartz, brass chips, iron ore, carbon black, and ceramics was developed by Deepika et al. (2013). Fig. 6 shows performance of developed and commercial pads under different inertia conditions. The average material loss per application of the developed pad, 4.2 mg, compares well with that of the commercial pad, 4.1 mg, and Blau (2001) reports a value of 3 mg for commercial brake pads.

Also, Elakhame et al. (2014) developed a novel brake pad using different particle size of PKS. The sieve PKS was used in a ratio of 20% resin, 10% graphite, 15% steel, 35–55% PKS, and 0–20% SiC using compression molding. The outcome of this study shows that samples that contain 100 μm of PKS gave better properties than other samples like 355 μm , 710 μm , and 1 mm size particles from the formulation. Moreover, hardness, compressive strength, porosity, and densities of the formed samples were decreased with the increase in sieve grade. Whereas, as sieve grade increased, the water soak, oil soak, wear rate, and percentage charred increased. Consequently, PKFs can be suitable replacement to asbestos for brake pad production.

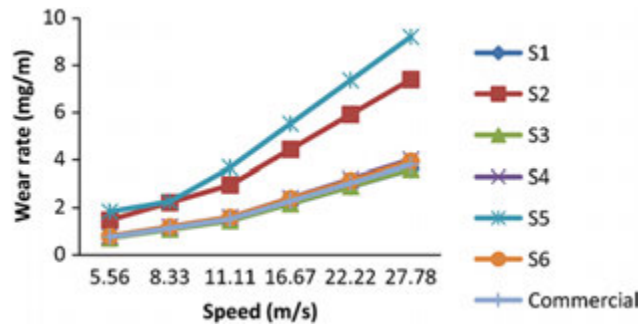


Fig. 4 Variation of wear rate with speed. Reproduced from Ikpambese, K.K., Gundu, D., Tuleun, L., 2014. Evaluation of palm kernel fibers (pkfs) for production of asbestos-free automotive brake pads. *Journal of King Saud University – Engineering Sciences* 28 (1), 110–118.

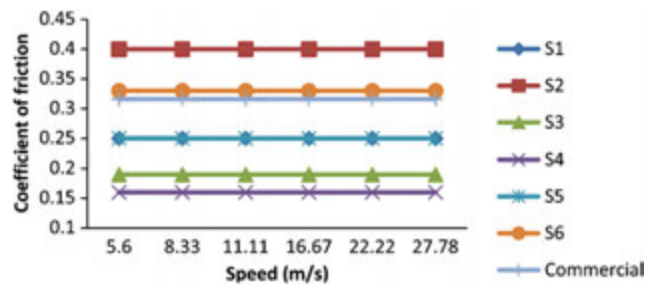


Fig. 5 Variation of coefficient of friction with speed. Reproduced from Ikpambese, K.K., Gundu, D., Tuleun, L., 2014. Evaluation of palm kernel fibers (pkfs) for production of asbestos-free automotive brake pads. *Journal of King Saud University – Engineering Sciences* 28 (1), 110–118.

Table 3 Summary of results compared with commercial brake pad composites

Brake pad-based	Wear rate (mg/m)	Coefficient of friction	Specific gravity (g/cm ³)	Thickness swells in water (%)	Thickness swells in SEA oil (%)	Hardness values (HRB)	Compressive strength (N/mm ²)	References
Commercial asbestos	3.80	0.3–0.4	2.06	1.18	0.39	101	110	Ikpambese <i>et al.</i> (2014)
Palm kernel shell	4.4	0.43	1.65	5.03	0.44	92.0	103.5	Ibhadode and Dagwa (2008)
Banana peels, carbonized	4.67	0.35	1.2	3.0	1.12	71.6	61.20	Idris <i>et al.</i> (2015)
Periwinkles shell particles		0.35–0.41	1.01	0.39	0.37	116.7	147	Idris <i>et al.</i> (2015)
PKS + CBS + MH	2.146	0.37–0.40	0.853	0.91	0.58	127.8	103	Ademoh and Olabisi (2015)
Maize husk	4.47	0.37	0.852	0.727	0.660	99.34	6.779	Ademoh and Olabisi (2015)
Bagasse	4.20	0.42	1.430	3.48	1.11	100.5	105.6	Aigbodion <i>et al.</i> (2010)
Coir	4.83		2.176			63.92	414.75	Maleque and Atiqah (2013b)
<i>Jatropha</i> seed cake	2.0	0.356	1.230			24		Shivamurthy <i>et al.</i> (2015)

Banana Peel Particles

Fabrication of a new friendly brake pad using banana peels was utilized by Idris *et al.* (2015). Morphology, physical, mechanical, and wear properties of the brake pad were studied. Results showed that compressive strength, hardness, and specific gravity of samples increased with increased resin; while the water soaks, oil soaks, wear rate, and percentage charred decreased as resin increased. Proper bonding was achieved with that of uncarbonized banana peel particles (BUNCp) at 20 wt% resin addition while that of carbonized banana peel particles (BCp) even at 30 wt% resin bonding has not been achieved to a high degree. In addition, compressive strength, specific gravity, and hardness of the produced samples were increased with the increase in wt% resin addition, while the water soak, oil soak, wear rate, and percentage charred decreased as wt% resin increased as shown in Fig. 7. Consequently, the samples, containing 25 wt% in BUNCp and 30 wt% BCp gave better properties in all.

Bashir *et al.* formulated a new brake pad material containing 13 ingredients, as shown in Fig. 8, including phenolic resin and banana peel powder as a modified binder (Bashir *et al.*, 2015). A reciprocating friction monitor is used to carry friction and wear

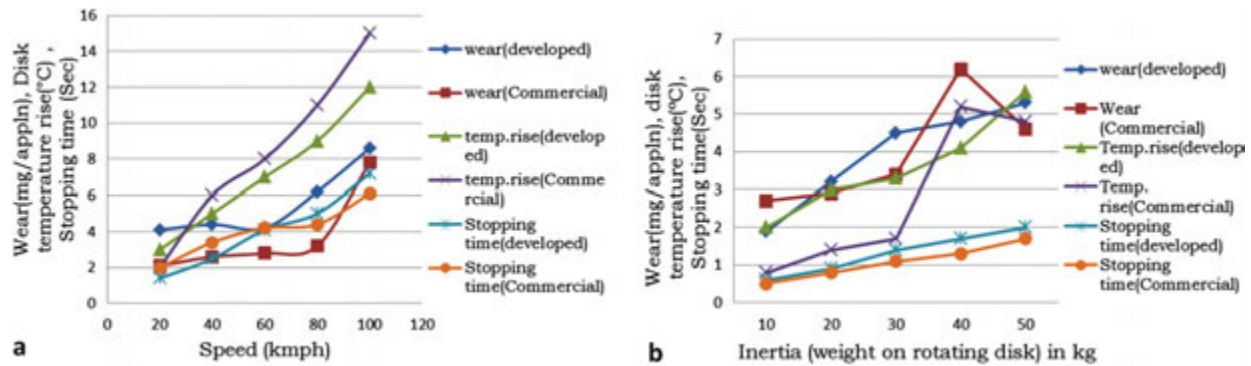


Fig. 6 Performance of the developed and commercial pads. (a) Performance under different inertia and (b) performance under speed condition. Reproduced from Deepika, K., Reddy, C.B., Reddy, D.R., 2013. Fabrication and performance evaluation of a composite material for wear resistance application. *International Journal of Engineering Science and Innovative Technology* 2, 66–71.

tests. Three tests, t1, t2, and t3, with different loads and temperatures were conducted for duration of 10 min. The results reported that the coefficient of friction increased at higher temperature and friction and wear characteristics indicate that banana peel powder can be effectively used to increase the binding ability of phenolic resin at higher temperature.

Sisal Fiber

The friction and wear characteristics of sisal fiber reinforced brake composites were tested using a constant speed tester by [Xin et al. \(2007\)](#). Different contents at separate friction temperatures were studied. The chemical and structural property of sisal fiber improved with modification, and was used to reinforce resin-based brake composites. Compared with the commercial disk brake pads, sisal fiber reinforced brake materials had optimal friction and wear properties when the proportion between resin and sisal fiber was 3:4. Sisal fiber S3 reinforced friction composites compared with asbestos C1, glass fiber C2, and mineral/steel fiber C3 are shown in [Fig. 9](#). It shows the good friction coefficient fitting with low wear rate at different friction temperatures. The results show clearly that the stability of friction coefficient of sisal brake material is the best. Such a stability of friction coefficient is one of the most important properties for brake applications.

Cedar Pine Cone (*Cedrus*)

Mutlu carried out brake pads using cedar (*Cedrus*) pine cone (CPC) dust along with boric acid (BA) ([Mutlu, 2009b](#)). Newly formulated brake lining materials with five different ingredients were tested under friction assessment and screening test (FAST). Experimental results have shown that the use of BA with the friction layer improved the general performance significantly. It simultaneously stabilized friction coefficient and faded reduction. Wear improvement was completed by the addition of BA and CPC dust to the formulation of the friction layer. The performance of the friction depends on the friction layer generated and differs from the formulation of the bulk material. However, it also depends on applied testing conditions and the environment. Finally less wear was observed with the samples where the BA rate is higher.

Rice Straw and Rice Husk Dust

Investigation of rice straw dust (RSD) and rice husk dust (RHD) as new materials to replace asbestos has started to be considered ([Mutlu, 2009a](#)). Composites were developed by using RSD or RHD and three ingredients including friction modifiers, space filler, and binder. The fabricated composites were labeled as RS4 and RS20 for RSD composites and RH4 and RH20 for RHD composites, respectively. RSD and RSH both shells have silica in them, which gives the pad materials a ceramic-like behavior. The experimental results have shown that the friction layer with the use of RHD improved the general performance significantly. In addition, wear rates were slightly increased to 20% of RSD and RHD and the best friction coefficient was achieved with 20% RHD ([Mutlu, 2009a](#)). Furthermore, friction surface and chemistry strongly affect the frictional performance on the friction layer; the structure of such friction layer differs from the bulk material formulation and also depends on the environmental and applied testing condition. [Fig. 10](#) shows the friction coefficient changes as a function of time.

Flax Fiber

A new brake friction composite that contains flax fibers was developed ([Fu et al., 2012](#); [Lu et al., 2010](#)). The composition consisted of mineral basalt fiber, plant flax fiber, and wollastonite as reinforcements; zircon as abrasive; natural graphite as solid lubricant; vermiculite and baryte as functional and space fillers; and cardanol-based benzoxazine-toughened phenolic resin as a binder. Flax fibers were isolated by chemical and physical methods and fibers with microfibrillated structure were formed on the surface. A new

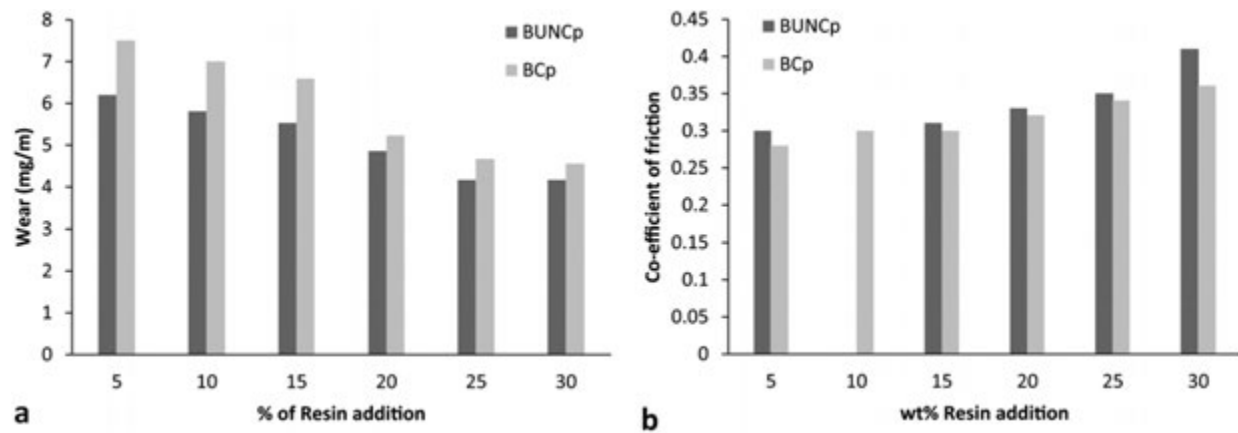


Fig. 7 Effect of resin addition on (a) wear and (b) coefficient of friction. Reproduced from Idris, U.D., Aigbodon, V.S., Abubakar, I.J., Nwoye, C.I., 2015. Eco-friendly asbestos free brake-pad: Using banana peels. *Journal of King Saud University – Engineering Sciences*, 27 (2), 185–192.

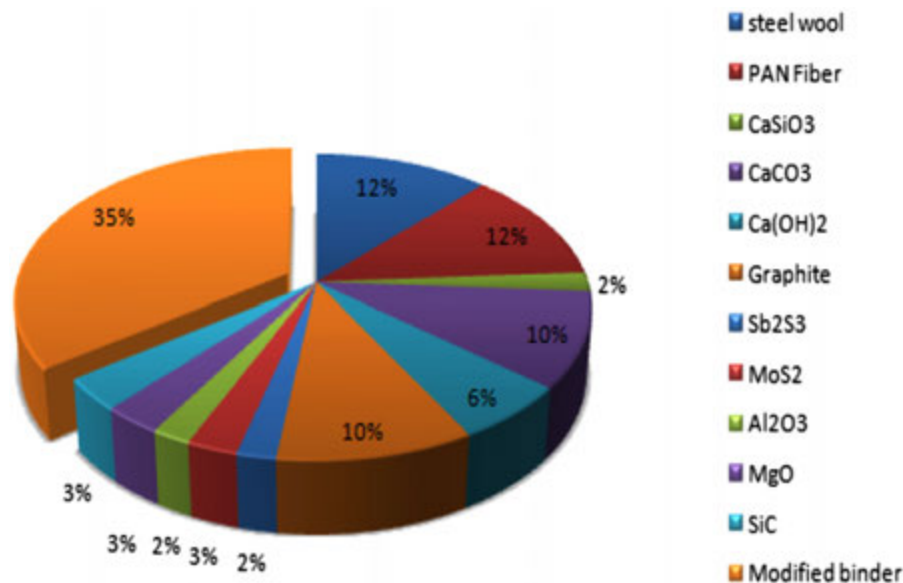


Fig. 8 Ingredients of the brake pad composite using banana peels. Reproduced from Bashir, M., Saleem, S.S., Bashir, O., 2015. Friction and wear behavior of disc brake pad material using banana peel powder. *International Journal of Research in Engineering and Technology* 4 (2), 650–659.

cardanol-based benzoxazine created by the reactions among aniline, cardanol, and formaldehyde was used as toughening for phenolic resin. The effects of both friction temperature and content of treated flax fibers on friction coefficient, friction performance, and specific wear rate of the friction composites were estimated by the method of extension evaluation. The friction sample containing flax fibers shows a slightly lower friction coefficient than the samples without flax fibers at lower temperatures, as shown in Fig. 11. The fade phenomenon appears on the friction coefficient under the effect of temperature. Hence, the ideal amount of flax fibers led to stable friction coefficient, which is one of the most significant demands for friction composites designed for automotive brake linings. Thus, the improved amount of flax fibers in the composites is 5.6 vol% (Fu *et al.*, 2012). Flax fibers play a big role in the composites, by increasing the wear rate, and stabilizing the friction coefficient at high temperature.

Pine Needle Fibers

Pine needle fibers were pretreated with alkali and mixed with other raw materials to produce pine needle fiber reinforced friction composites using compression molding to produce new eco-friendly brake pads (Ma *et al.*, 2014). The effects of pine needle fiber content were tested on the friction composites' tribological properties using a friction material tester at constant speed. The results revealed that the friction coefficient of the composites reinforced by pine needle fiber was very stable and markedly fade was not obvious compared with specimen FCO (containing 0 wt% pine needle fibers); the friction composites' wear rates usually increased when the temperature rose and were pointedly inclined by the test temperature. Results show that with the increase of the pine needle fiber content, the densities of the

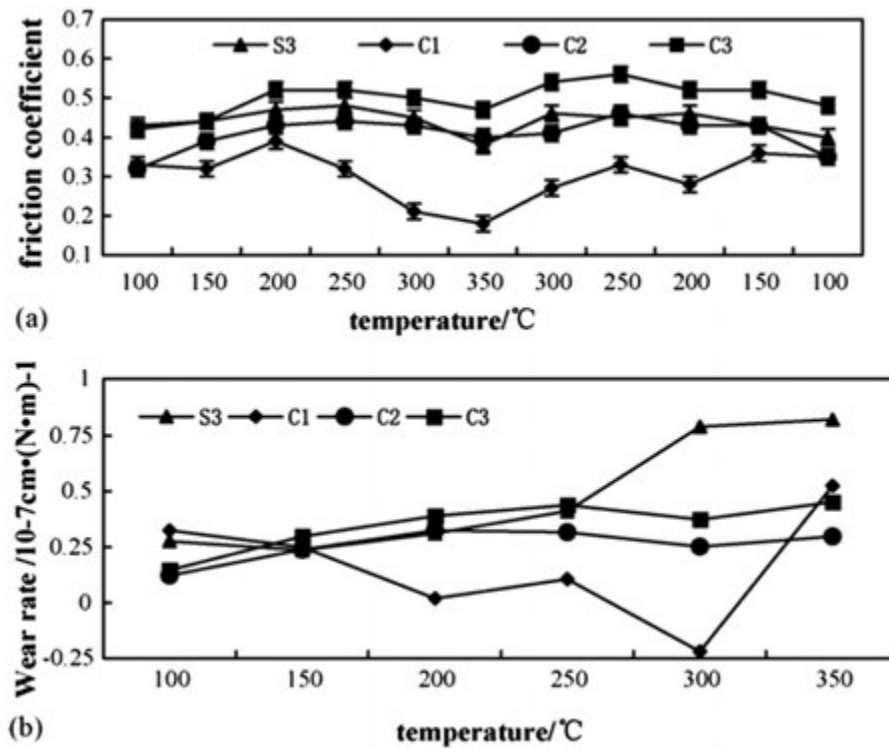


Fig. 9 Friction properties of brake materials reinforced with different fibers, (a) friction coefficient–temperature curves and (b) wear rate–temperature curves. Reproduced from Xin, X., Xu, C.G., Qing, L.F., 2007. Friction properties of sisal fibre reinforced resin brake composites. *Wear* 262, 736–741.

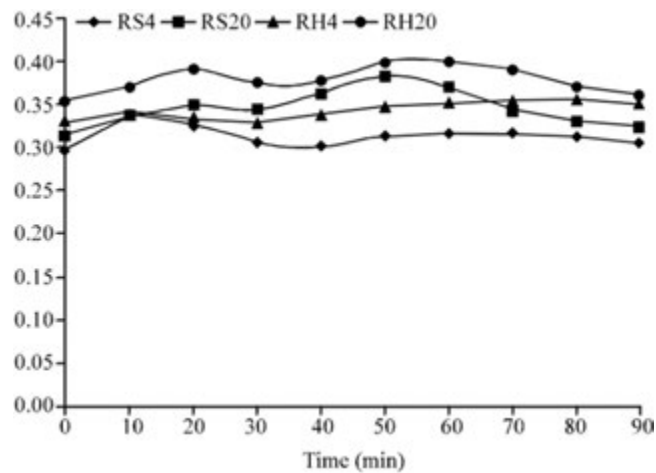


Fig. 10 Change of friction coefficient as a function of time for all samples. Reproduced from Mutlu, I., 2009a. Investigation of tribological properties of brake pads by using rice straw and rice husk dust. *Journal of Applied Sciences* 9 (2), 377–381.

friction composites decreased. Similarly, as shown in Fig. 12, the wear rates of the friction composites normally increased with temperature rise and a certain amount of pine needle fiber can obviously increase the interface bond between substrate and fibers.

Coir Fiber

Five different laboratory formulations, S1, S2, S3, S4, and S5, were prepared with variable coir fiber content of 0, 5, 10, 15, and 20% volume fraction respectively to develop brake friction composites (Maleque and Atiqah, 2013). Beside natural fiber the composite included friction modifier, binder, solid lubricant, and abrasive materials by using the techniques of powder metallurgy. The result shows that higher density, lower porosity, higher compressive strength, as shown in Fig. 13, and lower wear loss, were obtained from the 5% volume fraction of coir fiber reinforced composite.

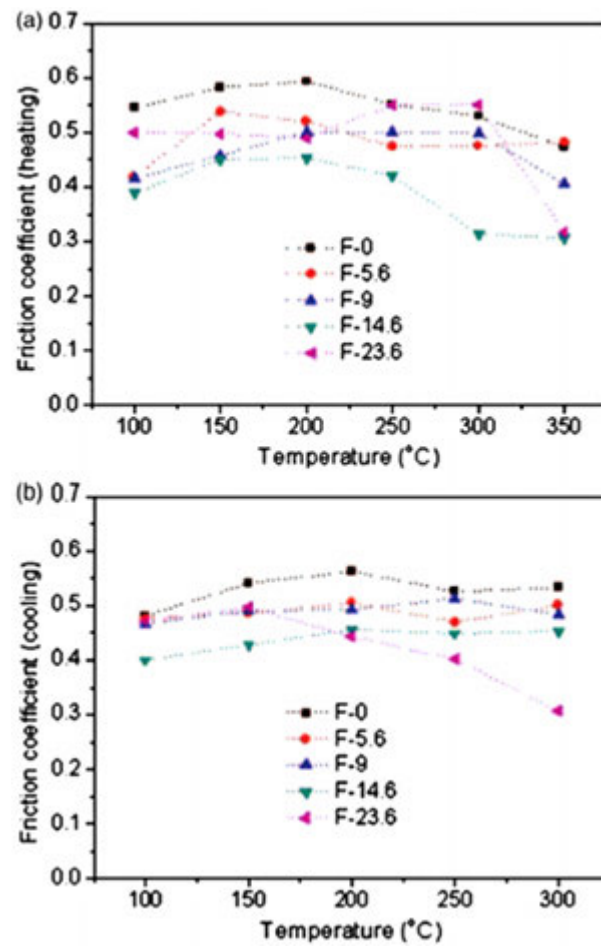


Fig. 11 Effects of flax content and temperature on friction measured during (a) heating process and (b) cooling process. Reproduced from Fu, Z., Suo, B., Yun, R., *et al.*, 2012. Development of eco-friendly brake friction composites containing flax fibers. *Journal of Reinforced Plastics and Composites* 31, 681–689.

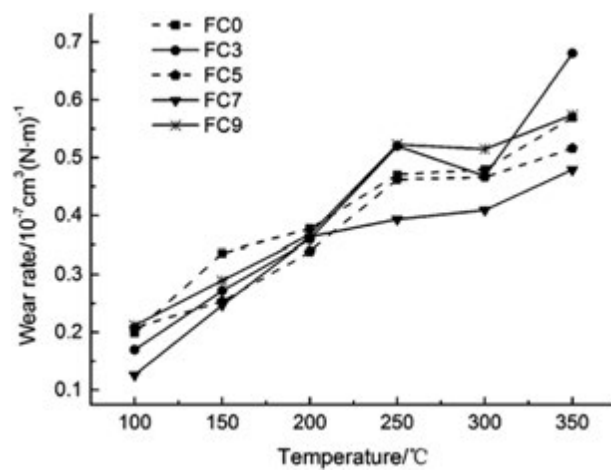


Fig. 12 Effect of temperature on wear rate. Reproduced from Ma, Y.H., Ma, S.S., Shen, S.L., Tong, J., Guo, L., 2014. Hybrid biological fiber-reinforced resin-based friction materials friction and wear performance test. *Applied Mechanics and Materials* 461, 388–396.

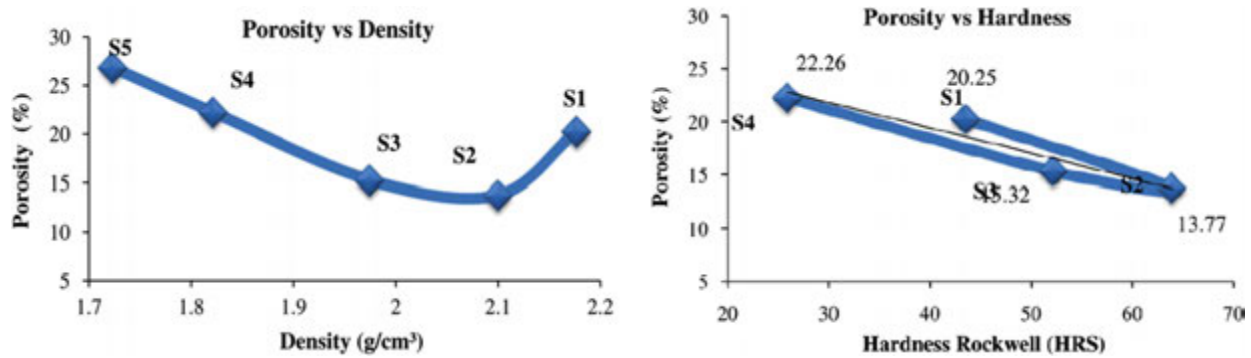


Fig. 13 Porosity vs. density and hardness of coir-based brake pads. Reproduced from Maleque, M., Atiqah, A., 2013. Development and characterization of coir fibre reinforced composite brake friction materials. *Arabian Journal for Science and Engineering* 38, 3191–3199.

Also, coconut fiber contents of four different laboratory formulations were changing from 0, 5, 10, to 15 volume fraction along with friction modifiers, binder, solid lubricant, and abrasive material using powder metallurgy method to develop new natural fiber reinforced aluminum composites (Maleque *et al.*, 2012). In addition to thermal properties, mechanical and physical properties were studied. The better properties with respect to lower porosity, higher compressive strength, and density were obtained from 5 and 10% coconut fiber composites. Moreover, Bashar *et al.* (2012) formulated a brake pad with multicomposite including epoxy resin binder matrix, ground coconut shell filler, iron chip reinforcements, cobalt naphthanate accelerator, methyl ethyl ketone peroxide catalyst, iron with silica abrasive, and brass as friction modifier. Both Bashar *et al.* (2012) and Maleque *et al.* (2012) stated that higher percentage coconut shell powder gave lower breaking, compressive, hardness, and impact strengths; implying that increasing the coconut powder increased pad brittleness.

Sugar Cane Fiber (Bagasse)

Development of an asbestos-free brake pad using bagasse was studied with a view to change the use of asbestos, the dust of which is hazardous (Aigbodion *et al.*, 2010). The bagasse was sieved into sieve grades of 100, 150, 250, 350, and 710 μm . Sieved bagasse was used to produce a brake pad in a ratio of 30% resin – 70% bagasse using compression molding. The examined properties are hardness, microstructure analysis, compressive strength, flame resistance, density, and oil and water absorption. The microstructure shows constant distribution of resin in the bagasse. Results revealed that the finer the sieve size the better the properties, as shown in Fig. 14.

Also, densities, compressive strength, and hardness of the fabricated samples were decreasing with the increase in sieve grade, whereas their water soaks, oil soaks, rate of wear, and percentage charred increased as sieve grade increased. The laboratory brake pads were tested for wear and effectiveness on a car. When compared with a premium asbestos-based commercial brake pad they were found to have performed satisfactorily. The results are in near agreement as shown in Table 3.

Hemp Fibers

A modified formula consisting of natural hemp fiber and environmentally friendly geopolymer as a fraction replacement for synthetic Kevlar fibers and phenolic resin, respectively, was reported by Lee and Filip (2013). The T-baseline sample consists of 3.4 wt% Kevlar fibers, 9.5 wt% phenolic resin, 3.6 wt% of antimony trisulfide, and 8.0 wt% copper. The modified samples consist of 0% of antimony trisulfide, 0% of copper, and 1.7 wt% hemp fibers. The T 403 and T 303 samples have 3.8 and 2.9 wt% of geopolymer, respectively. The Dyno result shows that the performance of the modified samples is better compared to the T-baseline when the temperature of the brake raised up in the fade section of the SAE 2430 test. The typical effectiveness in the fade section of T303, T403, and T-baseline are 0.41, 0.37, and 0.33, respectively. Yet, the modified samples reveal greater wear rate than the T-baseline. The T-baseline has 0.37 mm thickness, whereas the T403 and T303 samples lost 2.36 mm and 1.43 mm thickness, respectively, as shown in Fig. 15.

In addition, researchers at the Sustainable Technologies Initiative (STI) in the United Kingdom have devised a way to produce brake pads that are more sustainable by the use of renewable, sustainable crops including hemp (Morley, 2007). The new means of production utilizes hemp in place of aramid fiber but at no degradation in performance and less impact on the environment at cheaper costs. The project is in conjunction with a new sustainable lubricant, called Enviro-Lube, produced by partner PBW Metal Products Ltd. The new material was designed to support in the Tibrake project and does not contain heavy metals.

Palm Slag Fibers

Brake pads with palm slag fillers were investigated by Ruzaidi *et al.* (2012). Different pressure levels were employed during preparing samples through compression molding. It was shown that brake pad samples prepared with 60 t of compression pressure resulted in the most desirable properties as shown in Table 4.

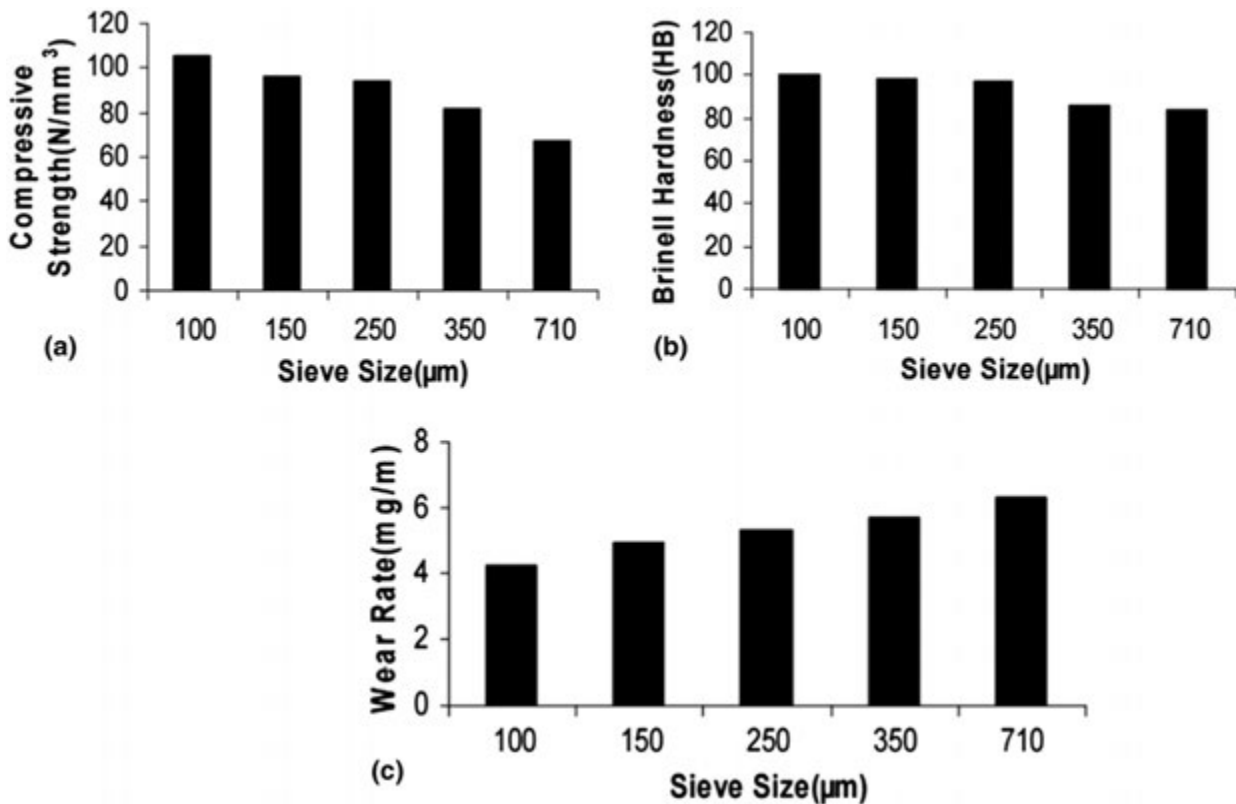


Fig. 14 Effects of sieve size on the properties: (a) compressive strength, (b) hardness, and (c) wear rate. Reproduced from Aigbodion, V., Akadike, U., Hassan, S., Asuke, F., Agunsoye, J., 2010. Development of asbestos-free brake pad using bagasse. *Tribology in Industry* 32, 12–18.

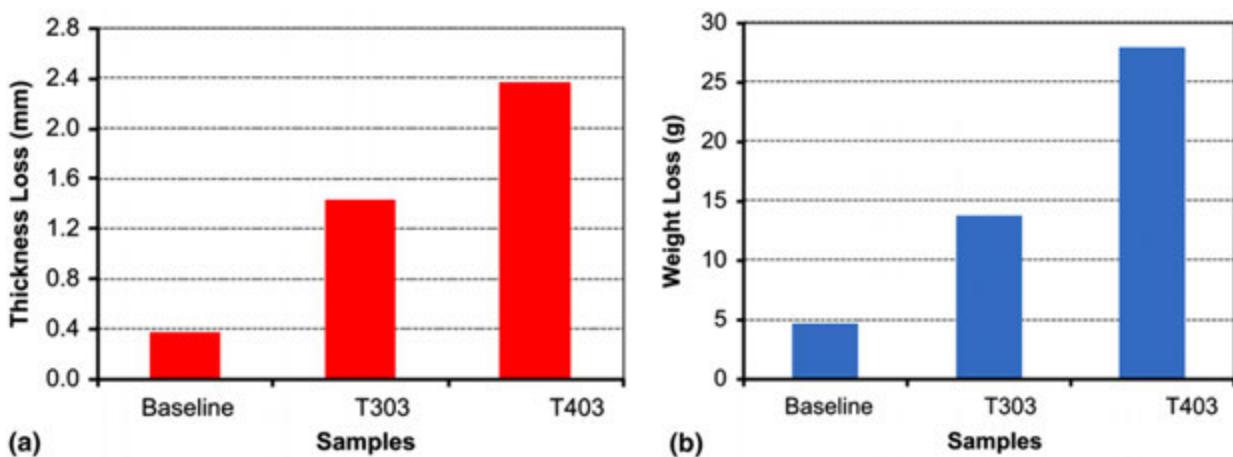


Fig. 15 The average (a) thickness and (b) weight loss of the tested samples after Dyno tests. Reproduced from Lee, P.W., Filip, P., 2013. Friction and wear of Cu-free and Sb-free environmental friendly automotive brake materials. *Wear* 302, 1404–1413.

Consequently, the processing, compression load plays an important role in enhancing the mechanical and wear properties of the product. The mechanical, thermal, and compressive properties of palm slag brake pad composite with other fillers are studies by [Ruzaidi et al. \(2011b\)](#). Moreover, mechanical and morphology studies were made to clarify the mechanism for compressive strength, hardness as shown in [Fig. 16 \(Ghazali et al., 2013\)](#). Also, the wear rate behavior of different filler of brake pad were investigated and prepared by compression molding of mixture of filler like palm slag, calcium carbonate, and dolomite in addition to phenolic as binder, metal fiber as reinforcement, graphite as lubricant, and alumina as abrasive. It was proven that palm slag has the potential to be utilized for brake pad composite as their wear rates were comparable with the conventional asbestos-based brake pad.

Table 4 Wear behavior of palm slag brake pad composite under sliding wear condition

Sample	Density (g/cm ³)	Wear volume (cm ³ × 10 ⁻³)	Wear rate (m ³ /m × 10 ⁻¹²)
Palm slag 10 t	1.93	2.02	2.02
Palm slag 20 t	1.98	1.77	1.77
Palm slag 40 t	2.01	1.09	1.09
Palm slag 60 t	2.02	0.89	0.89
Asbestos	2.22	0.72	0.72

Source: Reproduced from Navin, C., Hashmi, S., Lomash, S., Naik, A., 2004. Development of asbestos free brake pad. Journal MC 85, 13–16 and Ruzaidi, C.M., Kamarudin, H., Shamsul, J.B., Abdullah, M.A., Rafiza, A.R., 2012. Mechanical properties and wear behavior of brake pads produced from palm slag. Advanced Materials Research 341, 26–30.

Bamboo Fibers

Constant speed friction tester is used to investigate the tribology behavior of the bamboo fiber-reinforced friction materials (BFRFMs) (Ma *et al.*, 2013). Results revealed that the coefficient of friction of BFRFMs (reinforced with 3, 6, and 9 wt% bamboo fibers) was greater than those of the non-BFRFM with identical components and process conditions throughout the temperature increasing method as shown in Fig. 17.

When temperature rises during the temperature increasing method, the friction coefficient of 12 wt% BFRFMs decreased, but variation appeared at 200°C and then it decreased again with temperature rise. The friction coefficients of FRFMs with 3 wt% bamboo fiber were bigger than that with other bamboo fiber contents. The particular wear rate of BFRFMs increased with the temperature rise during the temperature increasing method, and the wear rates of BFRFM with 3 wt% fiber were lower than that of others. The friction coefficient of the friction material without bamboo fibers was lower than that the friction material filled with bamboo fibers. Fig. 18 shows that the voids or grooves formed after carbonization of the fibers reduced the noise and adhere to wear debris on the wear surface of the friction materials.

Scanning electron microscopy was used to analyze the morphologies of wear surfaces of friction materials (Eddoumy *et al.*, 2013). Results revealed that untreated bamboo fiber can reduce the noise and the particular wear rate and offer constant friction coefficient.

Maize Husk Fibers

The development and evaluation of maize husks as asbestos-free friction material for the production of automotive brake pad was carried out by Ademoh and Olabisi (2015). Three sets of composite compositions were made using maize husks as filler material to impart friction properties with varied epoxy resin contents as the matrix that bonded the particles in the mix. The particulate size of the MH filler material was 300 µm and epoxy resin was in slurry. The result showed that specimen composite 3 with 30% MH filler content having coefficient of friction, abrasion resistance, water absorption, oil absorption, density, hardness, tensile strength, compressive strength, and thermal conductivity of 0.37, 4.470E-6 g/m, 0.725%, 0.660%, 0.852 g/cm³, 99.34 MPa, 4.407 MPa, 6.779 MPa, and 0.330 W/mk, respectively, was optimum in performance.

It was observed that reducing the filler content increased hardness, wear rate as shown in Fig. 19, tensile strength, compressive strength, and thermal conductivity of the composite brake pad, while density, coefficient of friction as shown in Fig. 20, water and oil absorption enhanced with increased MH filler content.

Hybrid Natural Wastes

Matějka *et al.* (2013) used powdered hazelnut shells and jute fibers as natural fillers to fabricate nonmetallic NAO friction composites. If the temperature during the second fade test rises more than 300°C, the friction-wear tests show the jute with graphite samples to reveal important fade phenomenon. However, even at temperatures higher than 300°C, the samples of the jute with hazelnut shells show more resistance to the fade phenomenon. According to the comparison of the general performance of the friction, the composite with 8.4 vol% of powdered hazelnut shells and 5.6 vol% of jute fibers (recorded as JH-5.6) was considered as the best set, as shown in Fig. 21. This composite consists of 14 vol% of biodegradable, natural, and renewable components.

In addition, evaluation and performance of eco-friendly brake pads using natural fiber (JMM) were studied (Yun *et al.*, 2010). The composite samples were designed as follows: A1 and A2 were commercial brake pad samples, which were used as a reference; B: low-metallic sample; C: NAO composite sample using natural fiber; D: modified sample from C in which the aramid pulp replaced by further natural fibers; E: modified sample from D in which the coke partly replaced by alumina. Friction and wear performances (brake effectiveness) depend on the friction layer that formed on the surface of the pad. Moreover, thermal stability of brake pads as detected by thermogravimetric analysis (TGA), as shown in Fig. 22, well corresponds with hot performance of brake linings.

The method of extension evaluation was used to rank the friction materials using multiparameter criteria, including cost of raw materials, friction, wear, thermal stability, and parameters of the brake effectiveness evaluation procedure (BEEP) calculation. Moreover, wood pulp, jute, sisal, cotton as organic fibers and metal fibers, mineral wools, glass fiber, silica fibers and ceramic fibers as inorganic fibers were selected to develop friction materials (Bartram, 1980). The composition contains a thermoset binder, the

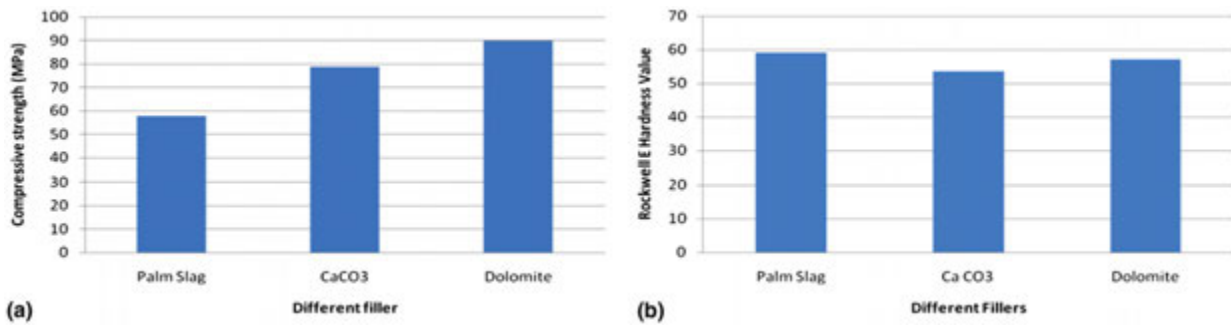


Fig. 16 Compressive strength and hardness of brake pad composite with different filler: (a) compressive strength and (b) rockwell hardness. Reproduced from Ghazali, R., Mohd, C., Kamarudin, H., *et al.*, 2013. Mechanical properties and morphology of palm slag, calcium carbonate and dolomite filler in brake pad composites. *Applied Mechanics and Materials* 313, 174–178.

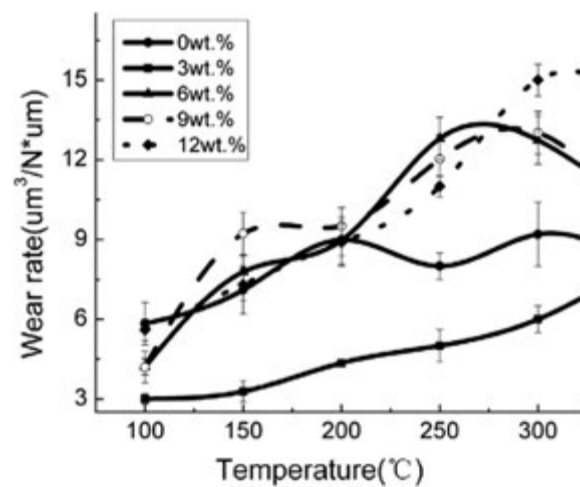


Fig. 17 Variation in the wear rate of the bamboo fiber load with temperature. Reproduced from Ma, Y., Shen, S., Tong, J., *et al.*, 2013. Effects of bamboo fibers on friction performance of friction materials. *Journal of Thermoplastic Composite Materials* 26, 845–859.

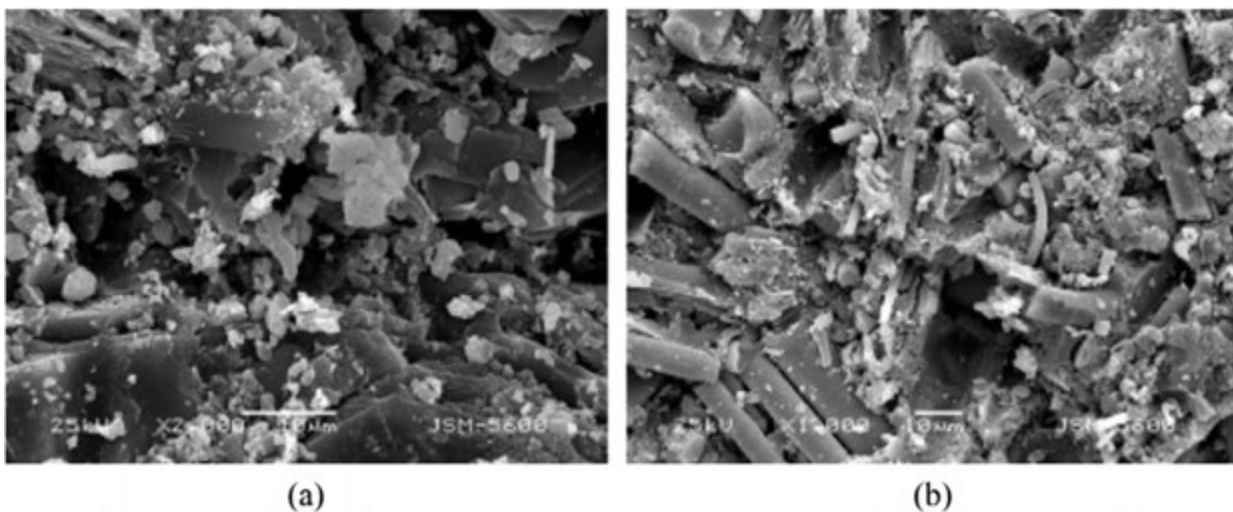


Fig. 18 Morphologies of the groove and void of the bamboo fiber-reinforced friction materials (BFRFs): (a) 3wt% and (b) 6 wt%. Reproduced from Ma, Y., Shen, S., Tong, J., *et al.*, 2013. Effects of bamboo fibers on friction performance of friction materials. *Journal of Thermoplastic Composite Materials* 26, 845–859.

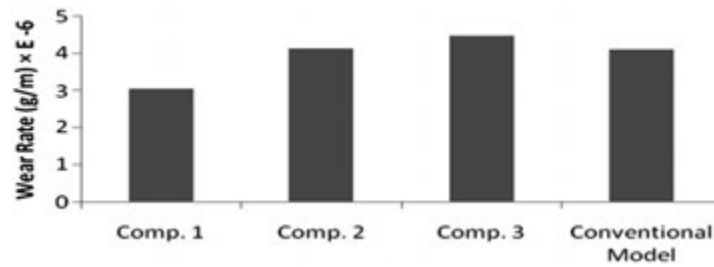


Fig. 19 Wear rate analysis of specimens. Reproduced from Ademoh, N.A., Olabisi, A.I., 2015. Development and evaluation of maize husks (asbestos-free) based brake pad. *Industrial Engineering Letters* 5, 67–80.

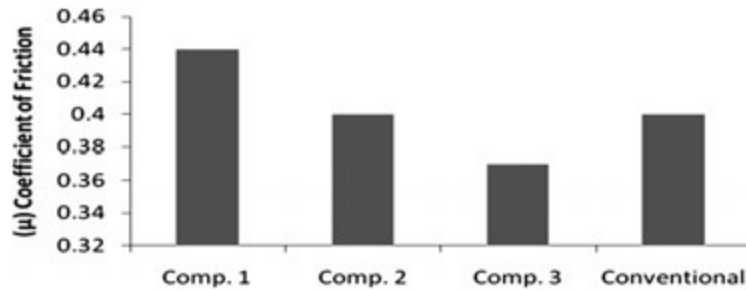


Fig. 20 Coefficient of friction analysis of specimens. Reproduced from Ademoh, N.A., Olabisi, A.I., 2015. Development and evaluation of maize husks (asbestos-free) based brake pad. *Industrial Engineering Letters* 5, 67–80.

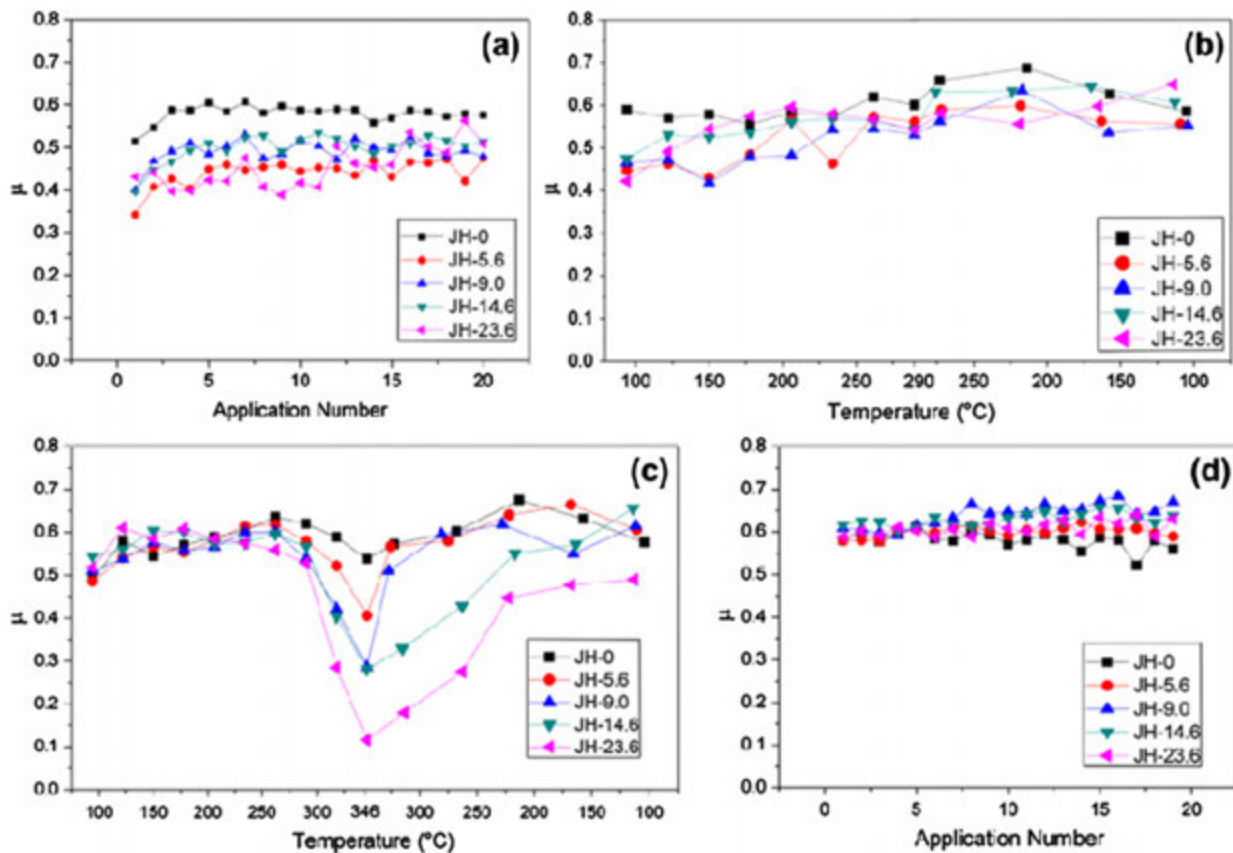


Fig. 21 Friction performances of the samples JH-X. (a) initial baseline, (b) first fade and recovery, (c) second fade and recovery and (d) final baseline. Reproduced from Matějka, V., Fu, Z., Kukutschová, J., *et al.*, 2013. Jute fibers and powdered hazelnut shells as natural fillers in non-asbestos organic non-metallic friction composites. *Materials & Design* 51, 847–853.

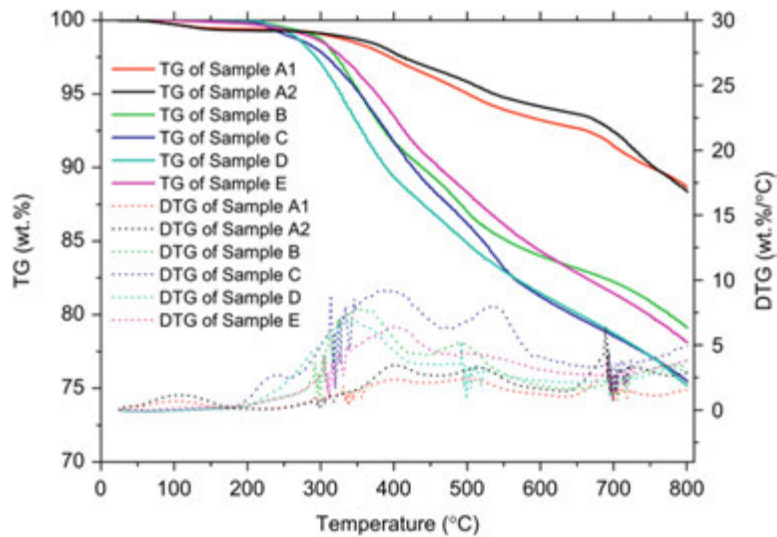


Fig. 22 Result of thermogravimetric experiment of all brake sample pads. Reproduced from Yun, R., Filip, P., Lu, Y., 2010. Performance and evaluation of eco-friendly brake friction materials. *Tribology International* 43, 2010–2019.

binder making up 15 to 40% by volume of the material. The fibrous reinforcement is a mixture of inorganic and organic fibers, which makes up about 70 to 33% by volume of the material.

Fibers of Nile roses displayed the highest friction values compared to the other tested natural fibers. Hardness of the brake pad has been controlled using suitable reinforcement materials like glass, carbon, and Kevlar pulp by Mathur *et al.* (2004).

The fabrication and design of a vehicle brake pad using commercially available materials in Nigeria have been effectively demonstrated by Onyeneke *et al.* (1998). The geometrical qualifications with a disk brake friction lining of Audi 90 model were formed using coconut shell and periwinkle shell as base materials. Epoxy resin and araldite were used as binder materials; aluminum, copper, and zinc were used as abrasives; carbon as fiber reinforcement; and rubber dust from shoes as filler. The experiment reported that the coefficient of friction of the pad material ranged from 0.4 to 0.65, wear rate of 0.025–0.06 mm/min, scratch hardness of 80–85, and bending strength of 25–27 kg/cm² as compared to the classic brake pad material, which has wear rate of 0.03–0.08 mm/min, hardness of 80–85, and bending strength of 25–27 kg/cm². These results agree with those of asbestos friction materials formed. Thus, it is recognized that this material could be applied in the Audi 90 model.

Eco-Friendly Brake Pads From Different Waste Materials

Periwinkle Shell Particles

Brake pads with different sieve size (710–125 μm) of periwinkle shell particles (PSP) with 35% resin were produced using compressive molding (Yawas *et al.*, 2013). Also, the coefficient friction of the composites decreases as the applied load increases, and will decrease with an increase in PSP size (Yakubu *et al.*, 2013).

Amaren *et al.* (2013) studied the same properties, but using factorial design of the experiment to describe the wear behavior of the samples, and developed a linear equation for predicting wear rate within selected experimental conditions. They found that the +125 μm particles of periwinkles gave the best wear resistance compared favorably with that of a commercial brake pad. Moreover, the wear rate increases with increase in sliding speed, load, and temperature. Besides, smaller particle size has a positive effect on the wear behavior of the samples as shown in Fig. 23. Also, as the periwinkle size increases, there is an increase in the interracial area; this increase results in poor interface bonding and in efficient stress transfer between particles and resin. The wear rates and the coefficients of friction reported by Amaren *et al.* (2013) are 75–125 mg and 0.35–0.41, while Yawas *et al.* (2013) reported 55–120 mg and 0.35–0.41, respectively.

Combustion Wastes

Combustion waste, specifically fly ash, consists of a mixture of fine-sized particles (mean particle size of 10–30 μm) of SiO₂, Al₂O₃, CaSO₄, and unburned carbon. These particles exhibit high-temperature resistance for breaking applications and provide good integrity/compatibility with the resin that dramatically enhances the friction and wear performance of their composites (Malhotra *et al.*, 2002). Particles of fly ash have certain features that make them appropriate to be used as a filler material in friction composites. An effort has been made to include more than 50 wt% of fly ash particles in the brake lining composites by Mohanty and Chugh (2007). They have developed friction composites, using fly ash from a certain power plant in Illinois. Components

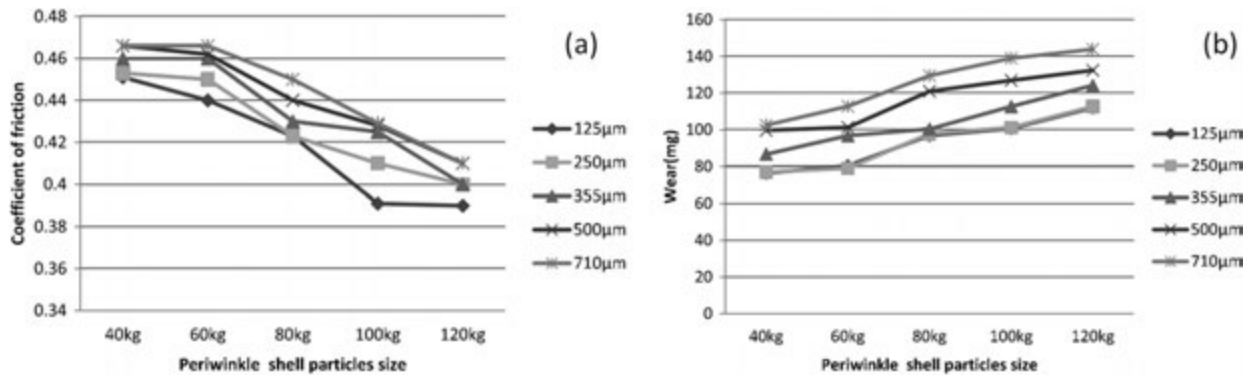


Fig. 23 Variation of coefficients of friction and wear with different sieve size at varying loads and constant speeds: (a) coefficient of friction and (b) wear. Reproduced from Yawas, D.S., Aku, S.Y., Amaren, S., 2013. Morphology and properties of periwinkle shell asbestos-free brake pad. Journal of King Saud University – Engineering Sciences.

such as aramid pulp, phenolic resin, glass fiber, graphite, potassium titanate, copper powder, and aluminum fiber were used in developing the composite, in addition to the fly ash. Developed brake lining composites showed wear rates lower than 12 wt% and reliable coefficients of friction in the range of 0.35–0.4. The friction performance of brake shoe composite indicated by coefficient of friction is influenced by braking conditions, including contact pressure, sliding speed, or temperature. This behavior is influenced by composite formulation. The results show that the coefficient of friction decreases with increasing volume fraction of phenolic resin and increases as the amount of fly ash is increased. In addition, phenolic resin affects load and speed sensitivity of coefficient of friction. In contrast, fly ash does not affect them.

Polychlorinated Biphenyl Wastes

A new brake pad produced by using palm ash and polychlorinated biphenyl (PCB) waste to replace asbestos together with metal filler and thermoset resin as a binder was introduced by Ruzaidi *et al.* (2011a). Four types of raw material are used, including palm ash, PCB waste, phenolic resin, and alumina. The composite that contains 35% palm ash, 35% PCB gives better wear properties. Compressive strength is increased with increasing the content of palm ash in the composition. Also, the sample with high content of palm ash gives better wear properties and lower water absorption, which results in better properties of the brake pad in application.

Current Challenges in Brake Pad Composites

In many countries, the gradual phasing-out of asbestos, copper, antimony, and other toxic materials in automotive brake friction materials has sparked the onset of extensive research and development into safer alternative materials. However, the design of the eco-friendly friction composites with the combination of waste of the natural plant fibers and other ingredients needs to be considered carefully as it was found here that brake pads have many challenges. That is, this systematic review revealed that brake pads made from natural wastes have lower thermal stability comparable to traditional ones, but contain much less toxicity and have much better performance from an environmental point of view. Moreover, the possibility of making green brake pads can be practical as the friction-wear performance of the composites could be significantly improved by selecting proper combinations of natural materials. In addition, most of the brake composites are usually a complex of different materials. These ingredients frequently comprise many disparate ingredients such as polymers, ceramics, and metals to satisfy several essential requirements such as wear resistance and high temperature friction stability under several functioning parameters such as applied loads, speeds, and temperature. Thus, some necessities have to be compromised to reach some other requirements. Generally, each design of friction material has its own unique wear-resistance characteristics and frictional behaviors. Moreover, the hybridization technique has been acknowledged as able to provide the balance between performance, cost, and more recently environmental attributes for natural fiber composites in many specific applications.

On the other hand, many factors should be considered for development of brake materials to fulfill requirements such as lower wear rate and stable friction coefficient at several operating speeds, temperatures, pressures, and environmental conditions in the automotive sectors. It is important to have an appropriate combination of materials in order to meet those requirements with reasonable cost of materials. The type and amount of these ingredients are determined mostly based on experience, empirical observation, or a trial and error method to make a new formulation. As conclusions from this work, several aspects have to be considered for green brake pads to achieve satisfactory results, like:

1. Study the tribology behavior of natural fibers in normal and hot conditions. These kinds of investigation will clarify the possibility of using natural fiber in friction composite (Ruzaidi *et al.*, 2011b).
2. Study the effect of fiber loads on tribology properties (Shalwan and Yousif, 2013; Amaren *et al.*, 2013).

3. Percentage in brake pad composite and their capability to withstand at evaluating temperature (Menezes *et al.*, 2012).
4. Further investigation of the mechanical and physical treatment to enhance the wettability, incompatibility of agro waste fibers with the matrix (Jawaid and Abdul Khalil, 2011; Koronis *et al.*, 2013).
5. Development novel brake pad composite-based waste materials as an alternative to the toxic ingredient of brake pad composites (Shivamurthy *et al.*, 2015; Al-Oqla *et al.*, 2015e).

Summary

It can be concluded that both mechanical and physical properties of the composites are influenced by the filler content. A decrease in filler content in composites increased hardness, tensile strength, compressive strength, and thermal conductivity of the composite brake pads, while density, water absorption, and oil absorption increased with increasing the filler content. Wear resistance improvements were achieved by adding natural fibers as fillers to the friction formulation in brake pad applications. Normal loads as well as speeds had significant impact on the specific wear rate of the brake pad composite. However, normal load had a more significant effect than speed on the composites. The wear rate increases with the increasing load and agro waste natural fiber loading. The performance of the friction mainly depends on the friction layer produced on the friction surface. Structure and morphology of the friction layer vary from bulk material formulation and depend on the environmental and test conditions applied. The small particle size of natural fibers has a positive effect on the wear rates of the composite. This may be attributed to the fact that as the particle size increases, there is an increase in the interfacial area bond between the particle and the resin, which reduces the possibility of a particle pull out, which may result in high wear. Consequently, the finer the sieve size, the more improved are the properties. The coefficients obtained were within the recommended standard for brake pad formulation. Physical and chemical treatments can be effectively used to overcome the poor wettability, incompatibility of natural fibers with high moisture absorption, and some polymeric matrices for braking pad applications. The agro waste might be efficiently used as a replacement for toxic ingredients in brake pad manufacturing when appropriately united with some other additives to fit a good performance of brake pad. On the other hand, regarding their low thermal stability, they should always be wisely considered.

Acknowledgments

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Life Cycle Analysis and Sustainability of Composite Materials: An Introduction

Lorna Fitzsimons, Advanced Processing Technology Research Centre, School of Mechanical and Manufacturing Engineering and the Water Institute, Dublin City University, Dublin, Ireland

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The focus of this section is environmental sustainability, and how this applies to composites specifically. The main focus areas are the methodologies that can be used to assess the environmental impacts of composites, waste strategies for composites, and manufacturing processes and energy management for composites. Environmental sustainability is defined in the Oxford English Dictionary as “the degree to which a process or enterprise is able to be maintained or continued while avoiding the long-term depletion of natural resources”. With regard to composites, the concept of environmental sustainability requires consideration throughout the full material and product life cycle. It relates to all resource flows, both inputs and outputs: materials, energy, chemicals, water, solid waste, wastewater, gaseous emissions. Life Cycle Analysis (LCA) of composites measures and quantifies the environmental impact of composites throughout the various life cycle phases for all of these aforementioned resource flows: the extraction, processing and refinement of raw materials; the manufacturing of composites, often using advanced processing techniques; the use phase of composites; and the end of life of composite materials, whether that involves reuse, recycling, recovery or disposal.

Composites are a combination of two or more materials, specifically functionalized to take advantage of their synergistic and superior engineering properties. The use and applications for composites are pervasive in society: aerospace, large structural components, electronics, sports goods etc. Their targeted performance characteristics can result in better environmental performance during their use phase. For example, lighter components with high specific strength used in lightweight vehicles can result in lower fuel requirements. Wind turbine blades made from carbon fiber composite materials can be lighter and longer, with increased fatigue performance, for increased power generation and longer service life. That said, in order to provide a more complete assessment of the overall environmental performance of composites, all other phases of the composites life cycle should be assessed. By definition, this should include all processing steps, the energy usage along the length of the product life-cycle, and the end of life options.

This section of the Encyclopedia includes several articles that focus on LCA. Several methodological challenges and limitations of LCA are discussed including goal and scope definition, functional unit selection, and boundary determination. The concept of Life Cycle Engineering is presented and applied in the context of composite materials. Two major topics relevant to the composites industry, namely the multifunctionality of composites and the reprocessing of composites are addressed. Another approach presented is the Material Design-for-eXcellence Framework, which addresses the challenge of evaluating the composite material's performance from a multi-dimensional property perspective, again underpinned by the LCA methodology.

One article focuses on additive manufacturing, and particularly the environmental sustainability benefits of additive manufacturing processes when coupled with the use of composites, for example, a reduction in material usage, reduced material wastage, reduced need for storage, and avoided obsolescence. Energy considerations for composites, addressed in a standalone article, includes not only the embedded energy in the materials (resulting from extraction and processing of raw materials), but also the electrical and thermal energy that is required for composites manufacturing. Effective energy management in manufacturing is therefore imperative to reduce environmental impacts.

Finally, the end of life for composites typically revolves around the reduce, reuse, recycle, reuse waste strategies. However, determining the optimum waste strategies for the various composite materials is not straightforward. These process options and major issues associated with composites are outlined.

Undertaking an LCA for Composites: Challenges and Limitations

Greg McNamara, School of Mechanical and Manufacturing Engineering, Dublin City University, Dublin, Ireland

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Introduction

Composite materials are currently used in a variety of applications, from the high-end automotive and aerospace industries, to renewable energy and domestic homeware products. The choice of composite material for a given application is often based on economics, but in today's climate with the heightened awareness for the need to produce more sustainable products and services, business leaders require more than just economic analysis tools. Life cycle assessment (LCA) is an analytical tool that facilitates a holistic approach to assessing the environmental performance of a product, process, system, or service from cradle-to-grave. A cradle-to-grave scope encapsulates all of the upstream and downstream process flows in a product's life cycle from ore extraction to material processing, manufacturing, use-phase, and the end-of-life (EoL) solution. The assessment tool began its development in the late 1960s as Resource and Environmental Profile Analysis (REPA) (Klöpper and Grahl, 2014). In 1997 the first of four ISO standards (ISO 14040) describing the *principles and framework for life cycle assessment* was published and since revised in 2006 (ISO, 2006). Since then, LCA has become the first globally accepted environmental assessment tool. However, it should be stated that LCA – like many analytical tools that produce results and recommendations that require interpretation and communication to a non-scientific audience – has its detractors. Many of the challenges and limitations experienced when conducting LCA studies of composite materials are similar to those experienced in other types of study; some of which will be discussed here, but most of which require a research-area specific review beyond the scope of this article. The structure of this article follows that of the general LCA methodological framework with discussion of many of the challenges encountered in particular phases, both in general and in relation to composite materials. To begin with, however, it would be useful to understand what is meant by the term 'composite material'.

Composites

It would be extremely rare nowadays to find a product that is entirely homogenous in its material composition except for maybe a solid wood piece of furniture, a steel column, or a glass bottle. Composites constitute a large percentage of the materials used in modern day manufacturing processes and construction projects. Exploitation of the combined effect of different material properties goes back as far as 3500 BCE. when the Mesopotamians would glue strips of wood together to make plywood, and later around 1500 BCE. the ancient Egyptians would use straw to reinforce mud bricks (Reddy Nagavally, 2017). Today, there are few areas of product development that do not use some form of composite either directly (in the product) or indirectly (in the production process). The common definition of a composite material - also known as 'composition material', or simply 'composite' – is that of a material which is produced from two or more materials with different physical or chemical properties; which, when combined, produce material properties different to those of the individual elements. The individual constituents of composite materials remain separate in the final structure and maintain their individual chemical and physical properties, which distinguishes them from solid solutions. Alloys are different to composites in that they consist of materials (metals and non-metals) that bond with each other at an atomic level, while the constituent materials in composites are physically and chemically different and no atomic bonding occurs. Vasiliev and Morozov (2013) make the important distinction that in addition to different properties, composites also have distinct boundaries between their constituent materials. These individual constituent regions should be large enough to be considered a continuum (Dawoud and Saleh, 2019). While there is a vast array of different composites, the most common structure is that of a reinforcement phase surrounded by a matrix or 'binder' phase. The use of steel reinforced concrete is commonplace in most civil engineering projects. While at a lower level, there is a wide selection of fiber-reinforced (glass and carbon) plastics used for a variety of applications. With such a broad selection of composite materials and products to choose from, and with extended producer responsibility (EPR) now a factor in modern production planning, LCA is considered by many in the manufacturing industry "as close to the gold standard of understanding the environmental consequences of a product as researchers can currently get." (British Plastics Federation, 2020). However, there are some areas of the methodology that present some unique challenges and warrant discussion.

Goal and Scope

The goal and scope of an LCA study is arguably the most important and difficult phase to get right, and is often revisited numerous times throughout a study as new data are collected and unearth unanticipated results. Here, the LCA practitioner and commissioner will agree on the terms of the study. The purpose, scale and scope, boundaries, functional unit, timeline, required resources, and intended audience of the study are determined during this phase. The purpose of a study (accounting, change orientated, or stand-alone) and intended audience (department level, company-wide, government, public) will dictate the level of complexity

involved, and the format for presenting results. Stand-alone audits, often used for marketing purposes or “hot spot” identification are relatively straightforward, in that there is no strict requirement to adhere to a common methodology (unless the purpose is for attainment of an ECO-label with predefined methodological assessment standards). Organizations can define their own standards internally, set their own benchmarks, interpret their own results how they see fit — a point often raised by LCA skeptics. However, for change-oriented assessments that involve comparative elements, the complexity increases because of difficulties that can occur when trying to achieve fair comparison between systems. Furthermore, comparative assessments that are intended for public consumption must adhere to the ISO 14040 series of standards to be considered a valid LCA study.

There are two types of LCA study being considered here as they pertain to composites: the study of the function of a product that has been manufactured from composites, and the study of the production of the material itself. There is an important distinction here and experienced LCA practitioners will be all too aware of the implications. The issue relates to the scope of a study. The ideal scope of any LCA study is cradle-to-grave, in which the impact of the entire life cycle is captured and assessed. In this case, a conventional LCA study that includes the entire life cycle from ore extraction to EoL can be undertaken if that is the desired scope. This type of study does not present many unique challenges outside of those normally encountered; however, there are some decisions regarding the choice of functional unit that should be given due consideration.

Functional Unit

In the LCA methodology, the functional unit is the reference flow to which all other flows and emissions are measured. In what is considered by many to be the very first LCA study, the Coca Cola Company commissioned a broad assessment of its packaging system, and wanted the study to account for energy, resources, and waste (Baumann and Tillman, 2004). The choice of functional unit in this case was straightforward because there was a single function - the transportation of a unit volume of beverage. There were several other desired *properties* for the potential container such as durability, stiffness, etc., but these should not be confused with the primary function of the container. Moreover, the options for alternative packaging being considered were three homogeneous materials (glass, polyethylene terephthalate (PET), and aluminum), which meant there were no attribution of function issues. This is an uncomplicated example for a simple product, but many modern composites are multi-functional and selecting an appropriate functional unit for an LCA study of a multi-functional system may not always be straightforward. For example, one could argue that a wastewater treatment system (WWTS) has a single function, and associated functional unit, namely, ‘the treatment of a unit volume of wastewater’. However, there are several processes involved in treating wastewater, many of which produce by-products or intermediate products, which themselves could be chosen as a functional unit, e.g., “the removal of a unit mass of sludge”, or “the production of a unit volume of biogas”. Because of the varied nature of wastewater composition, the use of “a unit volume of wastewater treated” as the functional unit may not always provide a fair comparison between systems (McNamara *et al.*, 2016). A system treating heavily polluted wastewater may have to consume more energy and resources, and will invariably produce more emissions than another system treating the same volume of wastewater but with less polluting constituents. For an example more pertinent to composites let us revisit beverage packaging; specifically, Tetra Pak packaging. Recall that packaging must have all of the necessary mechanical properties for transportation and maintenance of packaging structure, but may also need to have adequate preservation and barrier properties for beverages with limited shelf lives. Tetra Pak containers generally have several layers of polyethylene, a layer of cardboard, and a layer of aluminum that provides protection from light and oxygen which increases product shelf life. In a comparative assessment with competing packaging, the functional unit can no longer simply be the “transportation of a unit volume of beverage”, but now must be expanded to include the preservation element of the packaging. This means that a competing product system (e.g., high density polyethylene (HDPE) packaging) would have to be expanded to include the environmental impact from production and distribution of additional produce that would spoil without the additional barrier properties of the Tetra Pak packaging. This was considered in the Nordic Tetra Pak study by Jelse *et al.* (2011), but was excluded due to the lack of available data. The issue of the selection of an adequate functional unit for multi-functional materials was also highlighted in Hischier *et al.* (2017) as the first of three main issues related to LCA studies of nano-enabled materials, but which also has relevance to nano-composites which will be discussed in a subsequent section.

In the second scenario whereby the focus of a study is the production of the composite itself, the study is limited to a cradle-to-gate scope because the intended use of the material is not defined. In general, when conducting an LCA study of an object or material without a clear intended function, the standard functional unit must be changed to a declared unit as specified in EN 15804 (CEN, 2013). Depending on the aims of the study, this may not present any problem, if say, the purpose of the study is a stand-alone audit; however, for comparative assessments the problem becomes more complex. The first question to ask is what exactly are we comparing? We are comparing the ecological impact from the production of the competing materials. But are we comparing like with like if the materials have different properties, and if they have similar properties what is the purpose of the assessment? Unless of course it is the same material being produced from two different processes, in which case it is the process system, and not the product system that is being assessed.

Studies with declared units have particular relevance to composite materials. Let us take the example of a comparative assessment of two sheets of glass with a declared unit of $x \text{ m}^2 \times y \text{ mm}$ toughened glass. Glass A is a regular sheet, and Glass B is a composite sheet with a Nickel Oxide (NiO) film on one side. Both sheets have approximately the same weight and toughness. The addition of the NiO film on the composite glass introduces several other functions such as light transmittance adjustment, glare reduction, and solar heat regulation (Chen *et al.*, 2014). However, the benefits of these functions cannot be accounted for in a

comparative assessment without knowledge of the intended use. The glass could be destined for a window in a house or for solar panel applications, which, in both cases would have definite ecological benefits, but which are unquantifiable without knowing the intended application. Comparative assessments of this nature need to be conducted with a cradle-to-grave scope that includes a functional unit in order to capture the actual benefits and limitations of the competing product systems.

Data Acquisition

Life cycle assessment in general, is a data intensive operation, and conducting a study requires the collection and analysis of large tranches of data at different stages of the life cycle. In the LCA methodology, the life cycle inventory (LCI) stage is the collection of all input and output flow quantities (energy, materials, waste, emissions) associated with a system. Compilation of the LCI is the most laborious and time consuming stage of an LCA (Jolliet *et al.*, 2015), and depending on the scale and complexity the system under study, there can be thousands of associated flows. Many commonly used composites have well established inventories for a selection of regions. Gursel (2014) argues that most published LCA studies of concrete suffer from “a lack of data reflecting variations in technological and regional nuances”. However, this issue is not exclusive to composites, and has been highlighted as a general limitation of the methodology (Lueddeckens *et al.*, 2020). Many European industrial associations involved in composite production such as PlasticsEurope (PlasticsEurope, 2021) and Eurofer (European Steel Association) (Eurofer, 2021) have embraced life cycle thinking and have begun compiling their own energy and material inventories. Many of the leading LCA software and database packages such as GaBi (Sphera, 2021), SimaPro (SimaPro, 2021), and ecoinvent (Ecoinvent, 2021) come with a selection of composite LCIs and their associated characterization factors (LCIAs) as standard and additional datasets can be purchased on request.

Impact Assessment

The LCI forms the basis for the subsequent life cycle impact assessment (LCIA) phase, which aims to quantify the potential impact to human health, the environment, and resource consumption. In the LCIA phase, the flows of the system are firstly classified to an impact category. For example, a process may produce CH₄ gas (methane) which is classified as being a contributor to global warming, or having global warming potential (GWP), measured in base units of CO₂. Process wastewater emissions that flow into water bodies in the ecosphere can be classified as having toxicity potential. After the classification phase, the emissions have to be characterized, which requires characterization factors. Returning to the methane example, which is characterized with a factor of 25 times the GWP of CO₂ (Klöpper and Grahl, 2014), i.e., 1 kg of CH₄ = 25 kg of CO₂ equivalent. Without predetermined characterization factors an LCA study would be either much more time consuming, or incomplete, and final recommendations would be based solely on the inventory analysis. Of course there is nothing wrong with this approach except that the study could not be characterized as a full LCA in accordance with the ISO standards. In fact, many of the earliest LCA studies were limited to analysis of the LCI, referred to as *Life Cycle Inventory studies* (Klöpper and Grahl, 2014). However, most recent studies will include an LCIA phase, which introduces a problem for the relatively new “nanocomposites”, or “nano-enabled” composite technology. Nanocomposites are currently used in a wide range of applications in areas such as the biomedical industry (Hasnain and Nayak, 2018), food packaging (Azeredo, 2009), water purification (Pandey *et al.*, 2017), and renewable energy (Liang *et al.*, 2011). Salieri *et al.* (2018) compiled a review of 92 LCA studies of nanomaterials to assess what, if any, progress has been made in addressing the shortcomings highlighted in the studies. Throughout the literature, the three areas of particular concern that were identified were:

- (1) Selection of an appropriate functional unit that accounts for the material’s multi-functionality
- (2) The lack of inputs and outputs LCI data
- (3) The lack of LCIA characterization factors

The third of these in particular was highlighted in Hischier *et al.* (2017) as posing a significant challenge. The concerns specifically relate to the fate of the nanoparticle emissions, the extent of receptor exposure, and the level of the ecotoxicity produced. It has been concluded that much more research is needed before we can truly understand the human and environmental impact of nanocomposites, and nano materials in general.

Other issues related to the LCIA phase are more general in nature. The first of these concerns the non-mandatory sub-phases such as normalization, grouping, weighting and valuation. Some of these sub-phases are subjective and opinion based and are viewed by some as unnecessary, or too easy to manipulate for one’s own interests. The other widely debated topic on impact assessment is whether or not the methodology should describe the potential impact or the real impact. The difference between the two relates to the extent of data that are required for each. The advantage of describing impact in terms of potential according to Baumann and Tillman (2004), is that “a general science based methodology can be employed”. This methodology has real value in comparative studies where a potential impact can provide a sufficient indicator for decision making. A methodology that attempts to determine the real impact requires much more data for each of the parameters encountered along the cause and effect chain as described by Potting and Hauschild (1997). Furthermore, when considering primary, secondary, and tertiary impacts, particularly for global impact categories, there is uncertainty as to the level of accuracy and transparency that can ever be achieved with “real” or “end point” impact assessments. There have been some suggestions that both sets of analysis could be presented in

one consistent framework together or in parallel, but it was argued that could lead to conflicting models being presented next to each other (Bare *et al.*, 2000).

Recycling and End of Life

End of life solutions for composites are of particular interest to original equipment manufacturers (OEMs) who, in addition to being responsible for the environmental impact throughout their product's life cycle, are also responsible for their disposal, recycling, or reuse. The scope of the extended producer responsibility (EPR) is being broadened in EU Member States (MS) beyond electrical and electronic goods, batteries and vehicles (many of which include reinforced plastics) to include a wider range of other products and materials (European Commission, 2019). Landfilling and incineration of composites have either been banned in MS across the EU, or incur such high tariffs as to be prohibitive in cost (Marsh, 2003). The EU's Waste Hierarchy (European Parliament and Council, 2008) from most to least preferred is:

- (1) Prevention;
- (2) Preparing for reuse;
- (3) Recycling;
- (4) Other recovery, e.g., energy recovery; and
- (5) Disposal.

Composite EoL solutions, as they pertain to LCA, are as complex as the range of composite materials and products are extensive. However, the aim of this article is not to assess composite EoL options, but to highlight the challenges involved in their environmental assessments. As the general consensus is that the treatment and disposal option is to be avoided, a brief discussion here is limited to the remaining options. Source reduction is without doubt the most effective way to reduce impact and also the easiest method to assess; however, given that the use of composites continues to grow in popularity, it is probably the least feasible option amongst those listed. Energy recovery assessments cannot really be carried out in isolation but must be a part of a larger product system assessment that includes recycling and recovery. Problems with recycling assessments generally center around open loop recycling and cascading. Cascading maximizes resource-use by reusing materials in products that create the most economic value over the life cycles of subsequent products, ending with – if feasible – energy production. A guiding principle of this approach is the assertion that energy recovery should only be considered after all higher-value products have been exhausted.

In the LCA methodology, allocation refers to *“the attribution of environmental burdens during the life cycle, for co-production, recycling and disposal”* (Klöpper and Grahl, 2014). Allocation problems generally fall under one of three scenarios:

- (1) Multiple inputs – single output. Example, wastewater treatment plants have multiple inputs or sources and one output (if we consider only the treated wastewater as the output)
- (2) Single input – multiple outputs. Example, the production of different fuel oil types from a single oil extraction process.
- (3) Open loop recycling. Where the product is continuously recycled into a different product until it reaches its final waste stage.

For comparative assessments ISO 14040 states that allocation should be avoided, which leaves system expansion as the only other viable option. The amount of data that is required to expand the system to include each of the subsequent processes can be overwhelming and may not even be practical to achieve. There are other issues associated with assessing the recycling route such as how to account for the lower quality in the recyclates, but one of the overriding issues as highlighted in the study by Leahy (2019) on EoL options for composite material wind turbine blades, is that the LCA component of any possible solution cannot be assessed in isolation. Technical, environmental, social, and economic feasibility have to be considered together as part of a multi-criteria decision making process, which means that the resultant hierarchy of LCA results may not correspond with the frontrunners as decided by the other criteria. There is a large body of work on the topic of multi-criterial decision making, multi-criteria modeling, life cycle thinking, and whole life cycle analysis that deals with many of the issues raised here. A good starting point would be the review by Mardani *et al.* (2015).

Conclusion

Many of the LCA methodological challenges and limitations discussed in this article apply to more than just composites. Challenges related to goal and scope definition, functional unit selection, and boundary determination are universal and require skill and experience to overcome. Issues more specific to composites, such as life cycle inventory compilation require some “buy-in” from industry, which, thanks to the current focus on producing sustainable goods and services, is underway in many industrial sectors. Ongoing research into the impact of nanocomposite emissions will hopefully lead to the development of more spatially, and temporally specific impact characterization factors allowing for more robust, reliable, and complete LCIA phase results. Assessments of EoL options for composites encounter many of the same challenges that other product systems face and should be dealt with on a case-by-case basis as there is no one size fits all solution. It is clear, however, that the advantages of the life cycle assessment approach outweigh the disadvantages or limitations of the methodology, and the on-going research in academia and uptake in industry can only help strengthen the practice.

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Life Cycle Engineering of Composite Materials

Jasmin Dönmez, Alexander Kaluza, Felipe Cerdas, and Christoph Herrmann, Chair of Sustainable Manufacturing and Life Cycle Engineering, Institute of Machine Tools and Production Technology (IWF), Technische Universität Braunschweig, Braunschweig, Germany

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Introduction to Composite Materials and Need for Life Cycle Engineering

Composites represent one of the most important classes of engineering materials due to their versatility in terms of properties and applications (Clyne and Hull, 2019). Based on common definitions, composite materials consist of two or more materials, whereby the composition shall enable better properties than the individual materials for a specific application. Typically, the separate materials retain their individual properties in spite of amalgamating them (Chawla, 2012).

Composite materials are characterized by their high strength and stiffness combined with low density, so that they enable to realize a mass reduction compared to products made of conventional engineering materials, such as steel or aluminum (Campbell, 2010). This enables a decrease in the mass-related energy demands of technical systems such as cars. Associated efficiency gains can lead to a mitigation of product-related environmental impacts and thus represent an eco-efficiency strategy in product development. Composite materials, especially fiber reinforced plastics, are thereby studied and utilized to replace plastics and metals (Fleischer *et al.*, 2018). Besides the reduction of mass, other advantageous features of composite materials include corrosion resistance, media resistance, fatigue strength, burn-through properties, adjustable directional dependence of mechanical properties due to fiber orientation, energy absorption and low-wear surfaces (Neitzel *et al.*, 2014; Fleischer *et al.*, 2018). Therefore, composites are applied in many industries, such as infrastructure, construction, industrial, transportation or sports and recreation (American Composites Manufacturers Association, 2020).

However, from a life cycle perspective, the steadily growing use of composite materials might have drawbacks compared to conventional engineering materials. For example, the increasing amount and current way of handling composite waste already has a negative impact on resource conservation and the environment, e.g., increased landfill (Naqvi *et al.*, 2018). In order to reduce environmental impacts and to avoid problem shifting a life cycle perspective is required. Such a perspective is enabled by life cycle engineering (LCE), which aims at supporting the development of sustainable products. The core of LCE is the life cycle assessment (LCA) methodology, which aims to quantify the environmental impact of products and processes over their entire life cycle. In this regard, Fig. 1 shows the influence of a composite material design on the environmental impact of a vehicle, using the example of glass fiber and carbon fiber reinforced plastics, as an alternative to a steel reference. Looking at the first life cycle stage, the raw materials extraction, semi-finished part and parts manufacturing, composite materials initially carry a higher environmental impact, even if a weight reduction compared to a reference is realized. Based on these impacts, the slope α represents the benefits resulting from a decreased energy consumption during use. The break-even point indicates the distance that must be driven with the vehicle in order to compensate for the initial additional load compared to high-strength steel with the benefits in

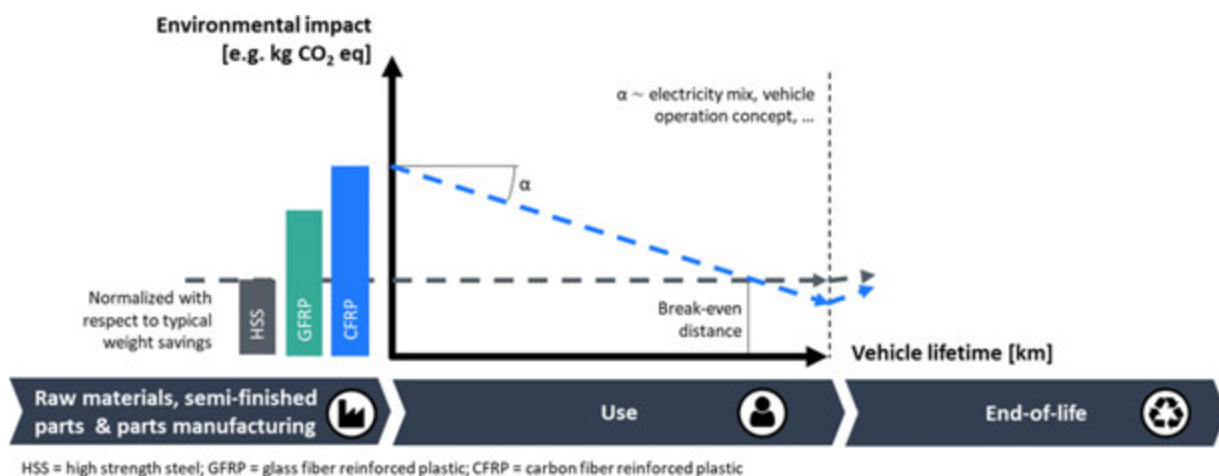


Fig. 1 Life cycle perspective of the influence of composites on vehicle environmental impacts. Based on Kaluza, A., Hagen, J.S., Dér, A., Marin, J.F.C., Herrmann, C., 2020. Life cycle assessment of thermoplastic and thermosetting composites. In: *Lightweight Polymer Composite Structures*, pp. 359–385. <https://doi.org/10.1201/9780429244087-13> and Dér, A., Gabrisch, C., Kaluza, A., *et al.*, 2019. Integrating environmental impact targets in early phases of production planning for lightweight structures. In *Procedia CIRP* 80, 168–173. doi:10.1016/j.procir.2019.01.077.

the use stage. The end-of-life includes impacts due to material recovery, avoided landfill and avoided primary material consumption (Kaluza *et al.*, 2017).

This article presents two current topics within the composites industry and relates them to engineering with regard to life cycle environmental impacts through LCA-based LCE. The following section will introduce core concepts of the LCE methodology. The subsequent sections further detail the environmental impacts of composite materials. This is followed by a LCA-based LCE perspective on multifunctional composite and on the role of composites in a circular economy.

Life Cycle Assessment-Based Life Cycle Engineering for Composites

In order to explore composite materials from an LCE perspective, the core steps of LCA-based LCE are presented in Section “Methodology” and then transferred to the field of composite materials in Section “Energy and Material Flows in the Life Cycle of Composite Materials”.

Methodology

Life Cycle Engineering (LCE) is an engineering approach that aims to mitigate negative environmental impacts of technical products over the entire life cycle (Herrmann *et al.*, 2014). LCE in a narrow sense aims to increase eco-efficiency of technical systems, meaning that the environmental impact per unit of function or cost should be minimized. The steady increase in population and affluence along with the associated environmental footprint, however, might mean that benefits from improved eco-efficiency are canceled out. Acknowledging the limited carrying capacity of the planet, there has been a shift from relative to absolute sustainability (Herrmann *et al.*, 2018). To align the scope of LCE to an absolute sustainability perspective, Hauschild and colleagues presented the Lyngby framework (Hauschild *et al.*, 2017; Kara *et al.*, 2018). In line with the framework, a new definition for LCE was introduced. LCE thus refers to “sustainability-oriented product development activities” (Hauschild *et al.*, 2017). The methods and tools supporting LCE aim to achieve an absolute reduction of the environmental impact of technologies. Population and affluence growth as well as the earth as a finite system are thereby considered (Bjørn *et al.*, 2015; Kara *et al.*, 2018).

As a systems analysis and assessment methodology, LCA enables the quantification of potential environmental impacts of product systems and processes (ISO 14044, 2006; Hauschild *et al.*, 2018). LCA is based on scientific principles and covers a wide range of environmental issues (Hauschild *et al.*, 2018). The fundamental principles as well as the framework of LCA are specified in various international standards (ISO 14044, 2006; ISO 14040, 2006). Based on the schematic diagram published by Herrmann *et al.* (2018), Fig. 2 shows an LCA-based framework of LCE, which is anchored in the ISO 14040 standards.

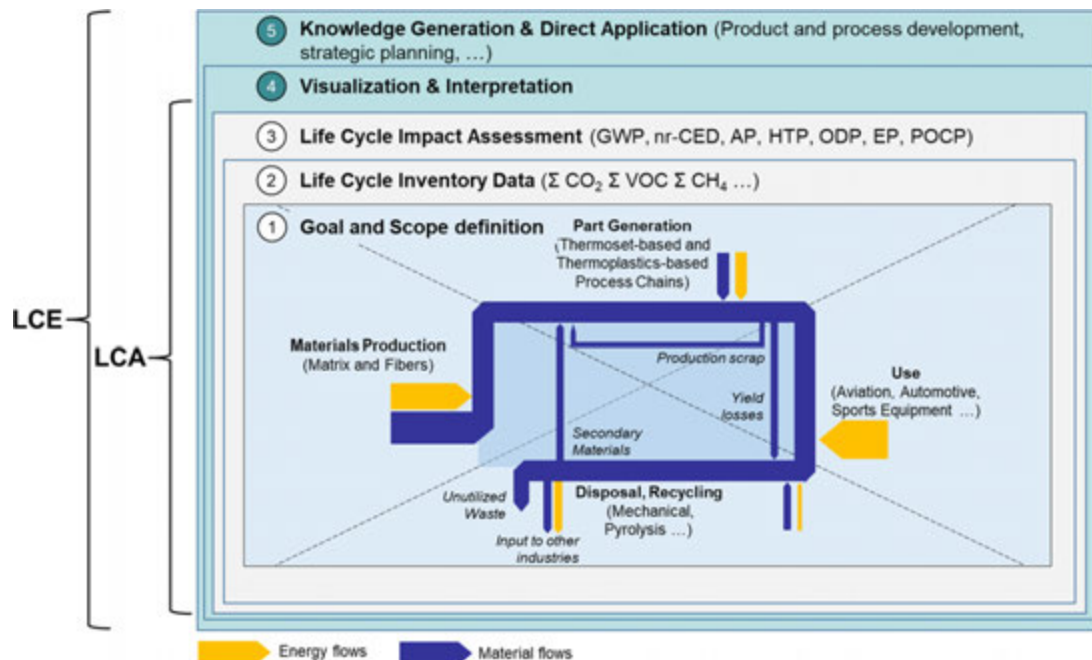


Fig. 2 Life cycle assessment (LCA) methodology according to ISO 14040 as part of the bottom-up life cycle engineering (LCE) methodology. Adapted from Herrmann, C., Dewulf, W., Hauschild, M., *et al.*, 2018. Life cycle engineering of lightweight structures. *CIRP Annals* 67 (2), 651–672. <https://doi.org/10.1016/j.cirp.2018.05.008>.

LCA-based engineering approaches can be applied to identify hotspots, avoid burden shifting and identify trade-offs. Furthermore, an understanding of the system can be gained and knowledge can be built. LCA-based LCE consists of five different phases and thus extends the ISO standardized LCA methodology by one further step (ISO 14044, 2006): (1) goal and scope definition, (2) life cycle inventory data (LCI), (3) life cycle impact assessment (LCIA) and (4) visualization and interpretation and (5) knowledge generation and direct application (Herrmann *et al.*, 2018). An iterative procedure in the individual phases characterizes the methodology. With the help of iterations, modeling decisions can be calibrated, e.g., when filling data gaps or performing consistency checks (EC-JRC, 2010).

- Within the first phase, the Goal and Scope Definition, the function and system boundaries of a specific application are determined. This comprises the definition of the functional unit, which represents the function provided by the product system under consideration, as well as the geographical, temporal and technological boundaries (Klöpper and Grahl, 2014; Hauschild *et al.*, 2018; Bjørn *et al.*, 2018). An LCA distinguishes between the foreground system and the background system. The foreground system thereby comprises all processes that are specific to the system under study and are thus in direct focus of the LCA study. The processes that are not specific to the system belong to the background system. Temporal and spatial differences in background systems can strongly influence the life cycle environmental impacts of composite materials. The types of environmental impacts intended to be investigated are also selected as part of the goal and scope definition (Hauschild *et al.*, 2018; Bjørn *et al.*, 2018).
- In the second phase, the LCI phase, all data required for the study is collected with the aim of creating a life cycle model. Such a model contains all energy and material flows across all processes of the product system. With regard to data collection, a distinction can be made for the foreground system between the collection of primary data, e.g., by means of measurements, and secondary data, from reports or scientific papers, among others (Lesage and Muller, 2017).
- The inventory obtained in the LCI phase is to be translated into an environmental profile (phase 3). This profile represents the impacts of the product system in specific impact categories (Hauschild and Huijbregts, 2015). The widely used midpoint indicators represent flows contributing to a specific environmental impact, e.g., greenhouse gas potential, measured in kg CO₂-eq.
- In phase four, the visualization and interpretation, the model itself as well as obtained results are evaluated. The life cycle model can be checked for completeness, sensitivity and consistency while the results can be analyzed in terms of hotspots or contributions. For complex product systems like composite structures, visualizations can support the decision-making process (Herrmann *et al.*, 2018).
- In the final phase of an LCA-based LCE methodology (see 5 in Fig. 2), knowledge is derived and directly applied to improve existing and future products and processes. To support management and engineering decisions, results of an LCA and knowledge derived from it must be available and accessible at the right time and place (Herrmann *et al.*, 2018).

Energy and Material Flows in the Life Cycle of Composite Materials

In order to determine an LCI for composites (phase 2 in Fig. 2), all material and energy flows over the entire life cycle must be taken into account. As a widely used and much researched type of composites, Fig. 2 exemplarily sets the life cycle of fiber-reinforced plastics (FRP) into the context of LCA-based LCE. In the center, the FRP life cycle stages with the corresponding material and energy flows are shown in the form of a Sankey diagram. The width of the arrow is thereby proportional to the quantity of the flow. The depicted stages include all processes that are directly linked or allocated to a FRP's life cycle. According to the description of Fleischer *et al.* four main stages are distinguished for the life cycle of fiber reinforced plastics (Fleischer *et al.*, 2018):

- **Materials production:** Fiber-reinforced plastics are composed of matrix and reinforcement materials. In the materials production stage, the fibers as well as the matrix materials are manufactured, which are then further processed to composite semi-finished products and parts. Different production routes can be distinguished with regard to the fibers and matrix materials (Kaluza *et al.*, 2020). Typical fiber materials are glass, aramid or carbon (Campbell, 2010; Fleischer *et al.*, 2018). Fiber reinforcements can be subdivided according to their length, which, in addition to the volume content and orientation of the fibers, is decisive for the component properties (Campbell, 2010; Fleischer *et al.*, 2018). Due to energy-intensive process steps, especially in the production of high-performance fibers such as carbon fibers, FRPs have a relatively high embodied energy (183–286 MJ/kg for carbon fiber, 72–112 MJ/kg for polypropylene compared to 30–60 MJ/kg for Steel) (Song *et al.*, 2009).
Polymers are widely used in industry as matrix material and can be further divided into thermoplastics, such as polypropylene or polyamide and thermosetting polymers, such as epoxy or polyester (Kaluza *et al.*, 2020). While thermoset matrix applications dominate the market for cost reasons, thermoplastic matrix systems are experiencing significant growth (Neitzel *et al.*, 2014). Process steps in the material production of thermoplastics and thermosets include the extraction of mineral oil, separation and refining, characterization and polymerization (Kaluza *et al.*, 2020).
Besides polymers, ceramics and metals are also commonly used as matrix materials in industrial applications (Fleischer *et al.*, 2018). In both, matrix and fiber production, bio-based and bio-degradable materials are gaining importance.
- **Part Generation:** This stage includes steps to obtain a functional composite product. Here a distinction can be made between processes that are suitable for thermoset or thermoplastic matrix systems or both. In addition, the production processes can be distinguished according to their technological maturity and realizable production quantities (Kaluza *et al.*, 2020). Manufacturing

processes include compression molding, liquid composite molding, injection molding, fiber deposition, thermoforming, pultrusion and filament winding (Fleischer *et al.*, 2018). The specific energy demands, required process materials and process emissions to air differ depending on the process (Kaluza *et al.*, 2020).

LCA studies for composite manufacturing chains are generally based on different scales of technology implementation, according to the market maturity of the process chains. Measured environmental impacts should therefore be analyzed in terms of possible technological improvements and thus an increase in market volume of the production technology under consideration (Kaluza *et al.*, 2020). Furthermore, high material yield losses also contribute to the production-related environmental effects (Hohmann *et al.*, 2017; Kaluza *et al.*, 2020). For example, in aircraft production it is estimated that 30%–50% of the materials are scrapped during manufacturing (Rybicka *et al.*, 2015; Kaluza *et al.*, 2020).

- Use: Since composite materials can be used for a wide range of applications in various industries, the use stage must be assessed for the specific product system under study. As part of a larger product system, a composite part or structure fulfils a specific function within that system (Herrmann *et al.*, 2018; Kaluza *et al.*, 2020). Within a life cycle assessment, the influence of a composite part on the overall function of the larger product system must be determined. As described in Section “Introduction to Composite Materials and Need for Life Cycle Engineering”, the introduction of a composite component as part of a lightweight strategy can reduce the energy demand in the use stage (see Fig. 1). The assessment of the use stage must be adjusted accordingly if the introduction of composite materials is not or only partially motivated by weight reduction (Kaluza *et al.*, 2020).
- Recycling & End-of-Life: Currently, most composite waste is being disposed of in landfills, which is increasingly restricted by legislation, such as the European end-of-life vehicle directive (2000/53/EC) (European Parliament and Council, 2000) and the directive on the landfill of waste (1999/31/EC) (European Council, 1999). Besides achieving compliance with legislative targets, treatment of production and end-of-life wastes is an important mitigation strategy for environmental impacts of composites. With regard to recycling and end-of-life treatment, composite materials present certain challenges, e.g., with regard to the separation of matrix and reinforcement material. Through different reprocessing strategies, such as recycling and remanufacturing, products or materials can be returned to the production stream and thus have a secondary use stage (Kaluza *et al.*, 2020).

Methodological Challenges in LCA-Based Life Cycle Engineering of Composite Materials

Composites are subject to rapid innovation cycles to improve products and processes along the life cycle. Among others, this includes works to leverage multifunctionality of composites as well as improvements in end-of-life. In their recent market analysis, Collins and Ghaffarzadeh refer to the concept of multifunctionality as the key next iteration for composite products (Collins and Ghaffarzadeh 2019).

Due to the constantly increasing amount of composite materials used, the treatment of composite waste streams is another important topic that industry and academia are dealing with. Current technological solutions lead to high degrees of landfilling, so that alternatives need to be investigated. In the following section, the multifunctionality of composite materials and the use of composite materials in closed loops will be discussed in more detail. LCA-based LCE can be leveraged to ensure the engineering of environmental benign products and processes. With regard to technological innovation, an LCE perspective should be adopted from the very beginning. However, the LCA-based LCE approach is not yet very widespread with regard to the assessment of multifunctional composites and composite materials in a circular economy. In the following, essential points and possible challenges in this context will be discussed.

Assessing Multifunctionality in Composites

Composites are ideally suited for implementing multifunctionality by combining the best features of different materials to realize a wide range of desired properties (Torquato *et al.*, 2002). However, the term multifunctionality in the context of composite materials is not clearly defined. Composites can be called multifunctional if at least one other function in addition to the primary structural function is fulfilled. Primary structural functions entail mechanical properties, such as stiffness, strength, fracture toughness, ductility, fatigue strength, energy absorption, damping, and thermal stability (Gibson, 2010; Ali and Andriyana, 2020; Narayana and Burela, 2018). Additional functions that can be realized by composites include electrical and thermal conductivity, sensing and actuation, energy harvesting/storage, self-healing capability, electromagnetic interference (EMI) shielding, recyclability and biodegradability (Gibson, 2010). Several authors reviewed the research area of multifunctional composites. Gibson (2010) points out advancements in polymer composites, nanomaterials and nanostructures. González *et al.* (2017) identify the integration of novel functionalities (mainly electrical conductivity and energy management) as key driver to enhance composites. Narayana and Burela (2018) stress the need to work on the mechanics of multifunctional composites.

Multifunctionality also plays an essential role with regard to LCE of composites, as efficiency and life span of a structure can be increased. Either more functionality can be realized with the same components or the same functionality can be realized with less components. This in turn can lead to a reduction in complexity, weight and cost as well as to an increase in material efficiency (Lincoln *et al.*, 2019). Compared to reference products, multifunctionality can enable lightweighting. In end-of-life, however, the integral design and the resulting heterogeneity of the components have a negative effect on possible treatment options. The separation into the individual constituents is difficult and currently leads to a high quality loss of the different materials. Therefore, it is essential to design multifunctional composite materials with the end-of-life and possible treatment options in mind (Composites UK, 2019). An integral design can also negatively affect the durability of products or components. If one component fails, this type of

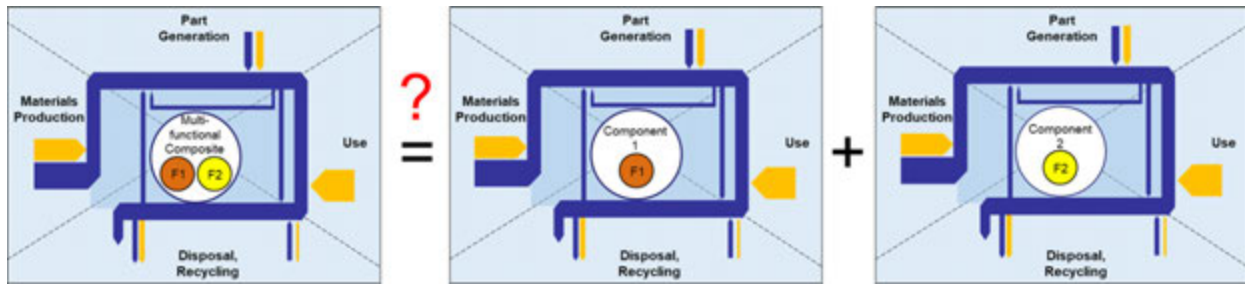


Fig. 3 System extension to consider the multi-functionality of composite materials, embedded in the life cycle schematic.



Fig. 4 Using system extension for the case study of Zackrisson *et al.* Reproduced from Zackrisson, M., Johannisson, C., Fransson, W., 2019. Prospective life cycle assessment of a structural battery. *Sustainability* 11 (20), 5679. <https://doi.org/10.3390/su11205679>.

design can result in the failure of the composite. In contrast, a modular design offers advantages such as the separate diagnosis of product components and the isolation of wear part offers, which is an important aspect for product maintenance (Bonvoisin *et al.*, 2016). A core question within LCA-based LCE is whether a multifunctional design is advantageous over a modular one. Therefore, it is important to understand the provided functions by the product systems under study because a comparison is only fair and meaningful if the compared systems provide (roughly) the same function(s) to the user (Hauschild *et al.*, 2018). Typically, a functional unit in LCA covers one function whereas for multifunctional product systems all provided functions need to be addressed. **Fig. 3** exemplarily shows a composite fulfilling two different functions, F1 and F2, as well as its comparison with unifunctional components embedded within the goal and scope definition. In order to deal properly with multifunctionality, LCA offers several approaches, which will be presented in the following paragraph.

ISO 14044 (2006) presents a hierarchy of solutions to handle multifunctionality. However, the standard focuses on processes shared by different products and does not address multifunctional products per se. The first priority is to avoid allocation in handling input and output flows. This can be achieved by subdividing the examined process until a clear allocation to a specific product system is possible. Alternatively, the product system can be extended by the additional functions of the co-product, requiring the adjustment of system boundaries accordingly. If allocation is unavoidable, the inputs and outputs of the system should be assigned according to underlying physical relationships or economic value of the co-products (ISO 14044, 2006). In LCE of multifunctional composites, a further breakdown into subfunctions would require extensive rules that would be difficult to obtain, as a sub-division of functions contradicts the basic character of composites. A system extension (**Fig. 3**) is, however, another option. The functional unit would therefore have to indicate exactly the specifications of the functions F1 and F2 of the multifunctional composite component, which is depicted on the left side of the equation. In order to enable a meaningful comparison, the system boundary must be expanded to also include different monofunctional components. However, it is difficult to take proper account of emergent functions that result from the integral character of composites (indicated by the question mark in **Fig. 3**).

Only a few publications deal with the environmental assessment of multifunctional composites. Zackrisson *et al.* (2019) performed an LCA of a structural battery built into the roof of an electric vehicle, capable of storing electrical energy and carrying mechanical loads. The structural battery thereby replaces the original steel roof of the vehicle and part of the original traction battery. In order to achieve a meaningful comparison, the authors performed a system expansion for replacing a steel roof and an equivalent part of the original traction battery with the structural battery roof (see **Fig. 4**) (Zackrisson *et al.*, 2019).

Composite Materials in a Circular Economy

A large part of composites is produced on the basis of crude oil. Further, composite products show a slow degradation at the end of the life and contribute to the increasing plastic contamination, resulting in a negative impact on resource conservation and the environment (Naqvi *et al.*, 2018). In order to further develop composites regarding sustainability targets, it is important to decouple economic growth and resource consumption and to find alternatives to the traditional linear “take-make-dispose” economy. The circular economy, in

which used resources are completely recirculated to different phases of the life cycle through various reprocessing strategies, represents such an alternative (Ellen MacArthur Foundation, 2013; European Commission, 2015). Strategies include durable design, maintenance, repair, reuse, remanufacturing, refurbishing and recycling (Geissdoerfer *et al.*, 2017). Composites can be transferred into several life cycles at product, component or material level, helping to reduce or avoid the need for virgin material (Fleischer *et al.*, 2018). Closing material and energy loops through reprocessing strategies is motivated by reducing environmental damage compared to the material and energy intensive processing of virgin materials. Nevertheless, the environmental impact associated with the circulation of used products, for instance caused by energy demand required for the separation of materials, cannot be neglected (Zink and Geyer, 2017). Further, due to the nature of composite materials, process steps such as sorting and separating in particular can be more energy-intensive compared to conventional materials (Herrmann *et al.*, 2018).

As the prevalent reprocessing strategy (Rybicka *et al.*, 2016), recycling of composite materials will hereafter be examined in the context of LCA-based LCE. In order to ensure that the recycling of composites does not negatively impact the environment, it is essential to carry out a complete evaluation to compare the different techniques in regard to environmental impact, efficiency and economic viability (Oliveux *et al.*, 2015). This needs to take into account quality considerations as well as time effects on material stocks (Herrmann *et al.*, 2018). Fig. 4 depicts the influencing factors that need to be considered to assess the net environmental impact of recycling. In order to achieve net environmental benefits, the impact of the reprocessing strategy E_{repro} , including all process steps such as the collection and separation, needs to be lower than the impact of the avoided primary production of raw material E_{prim} as well as the impact of the avoided landfilling activities E_{landfill} (Geyer *et al.*, 2016).

Collection and Reprocessing: The recycling of composite materials presents various challenges. The complex composition of fibers, matrix and fillers, makes separation difficult while the cross-linked character of thermoset resins inhibits remolding. Further, composite materials are often combined with other materials (Pimenta and Pinho, 2011), e.g., as sandwich materials. As a result, increased shares of composites might reduce material recovery rates and lower the quality of recycled materials (Herrmann *et al.*, 2018).

The separation of the reinforcement (fiber/filler) from the matrix components is a basic requirement for recycling composite materials. Different techniques can be distinguished. For glass and carbon fibers, these include biotechnological, chemical, electrochemical, fluidized bed, high voltage fragmentation, mechanical, microwave pyrolysis and pyrolysis (Mativenga *et al.*, 2016; Fleischer *et al.*, 2018). Mechanical treatment is the most mature technique for glass fiber composites, while pyrolysis is the most mature one for carbon fiber-reinforced composites (Rybicka *et al.*, 2016). All techniques differ in terms of energy intensity and quality of the recovered fiber and matrix (Fleischer *et al.*, 2018). In mechanical recycling recovered fibers are typically shorter than virgin fibers, leading to reduced mechanical properties and thus low-value secondary applications. Also in pyrolysis, the recovered fibers are of lower quality than virgin fibers due to insufficient surface properties. However, it was found that the quality conditions of recycled fibers correlate strongly with the pyrolysis conditions (Pimenta and Pinho, 2011).

Avoided primary production: Composite materials have a comparatively high embodied energy, which is mainly due to the energy-intensive fiber production (Fleischer *et al.*, 2018). The reprocessing of such materials should be less energy-intensive than their primary production to deliver environmental benefits (Geyer *et al.*, 2016).

Avoided landfill: Landfilling is the least favorable alternative in the waste hierarchy published in the European directive 2008/98/EC (European Parliament and Council, 2008). This hierarchy indicates an order of priority with regard to the environmental impact, establishing waste prevention as the best option. Reusing products comes in second place, followed by recycling (European Parliament and Council, 2008). From a technical point of view, the disposal through landfilling would not leverage the high embodied energy required to manufacture composite parts (Song *et al.*, 2009). Especially in regard to carbon fiber there is also an economic incentive to recover the remaining value due to the price of the virgin fibers (Howarth *et al.*, 2014). In this context the mixture complexity also plays a role. Products with a low mixture complexity and high amounts of valuable materials are recycled in contrast to those with a higher mixture complexity and a lower concentration of the material (Dahmus and Gutowski, 2007).

Material markets: To reduce the environmental impact in absolute terms, the displacement of virgin raw material production through secondary material plays an essential role (Geyer *et al.*, 2016). In this regard, several authors emphasize the necessity to investigate the dynamics of demand and supply of the material markets, to develop a respective secondary market as well as to elaborate possible fields of application (Geyer *et al.*, 2016; Meng *et al.*, 2018; Kaluza *et al.*, 2020).

To determine whether the circulation of composite materials is environmentally benign a respective assessment should be carried out based on the previously introduced framework. The circulation of material and energy is also addressed by ISO 14044 (2006). Geyer *et al.* (2016) point out that ISO 14044 does not address market dynamics but instead focuses only on the inherent material properties of the recycled material. If the material properties of the recycled material remain the same, it is assumed that virgin material will be displaced by secondary material. This kind of one-to-one displacement is, however, only an assumption and cannot be considered as a fact (Geyer *et al.*, 2016). In LCA of the recycling of composite materials, it is therefore essential to know the actual quantities displaced in order to avoid a systematic overestimation, which could be the case if a one-to-one displacement is assumed for all recycled materials where the inherent properties have not changed (Geyer *et al.*, 2016).

For the LCA of recycling allocation procedures are also applicable. Allocation might be required both for the evaluation of product systems whose inherent material properties have been changed by recycling and for recycling processes that are shared by several product systems. It should be based on physical properties, economic value or the number of subsequent uses of the recycled material (ISO 14044, 2006). If the material properties remain the same and thus a one-to-one displacement can be assumed according to ISO, allocation can be avoided. Instead, the assessment can be done by a system extension where the process

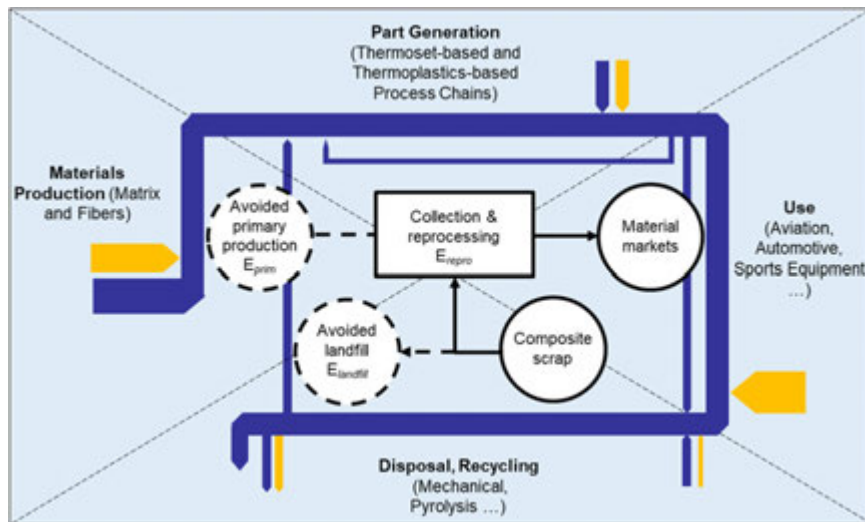


Fig. 5 Influencing factors to be considered for assessing the environmental impacts of recycling integrated into the composite life cycle. Adapted from Geyer, R., Kuczenski, B., Zink, T., Henderson, A., 2016. Common misconceptions about recycling. *Journal of Industrial Ecology* 20 (5), pp. 1010–1017. <https://doi.org/10.1111/jiec.12355>.

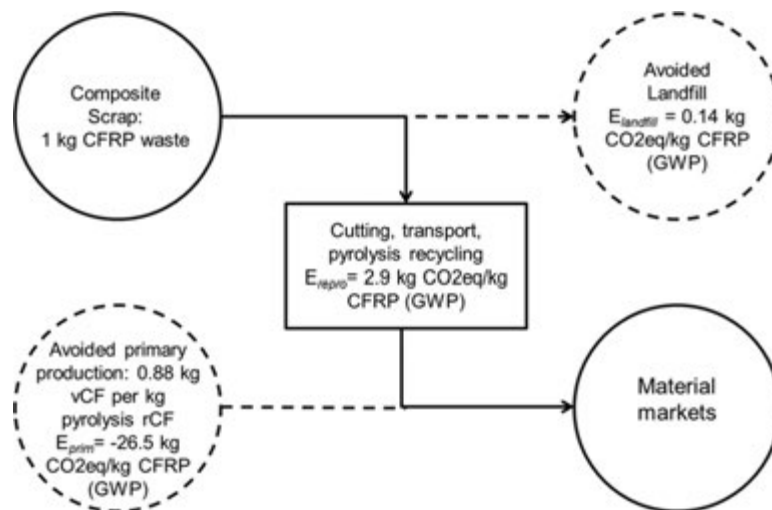


Fig. 6 Influencing factors to be considered for assessing the environmental impacts of pyrolysis recycling. Based on Meng, F., Olivetti, E.A., Zhao, Y., *et al.*, 2018. Comparing life cycle energy and global warming potential of carbon fiber composite recycling technologies and waste management options. *ACS Sustainable Chemistry & Engineering* 6 (8), 9854–9865. <https://doi.org/10.1021/acssuschemeng.8b01026> and Geyer, R., Kuczenski, B., Zink, T., Henderson, A., 2016. Common misconceptions about recycling. *Journal of Industrial Ecology* 20 (5), pp. 1010–1017. <https://doi.org/10.1111/jiec.12355>.

inventories of the recycling process are added and then the inventories to produce an equal amount of primary material subtracted (ISO 14044, 2006; Geyer *et al.*, 2016).

Also Hauschild *et al.* (2018) present weaknesses within the current practice of LCA with regard to recycling. In order to enable an adequate assessment of recycling processes and recycled material, the representation of the material's constituents within the LCI modeling practice must be improved. Besides intended heterogeneity, as in the case of composite materials, unintended heterogeneity, e.g., due to impurities in the use phase, should be taken into account. It must also be considered how heterogeneity affects the recyclability of the material. Furthermore, the composition of the recycled materials must also be included. In order to consider multiple life cycles in LCI modeling, Niero and Olsen (2016) suggest that the functional unit includes all functions that a material performs in the planned life cycles rather than just the function in the first life cycle (Hauschild *et al.*, 2018).

The state of research broadly covers the recycling of carbon fibers. Although the amount of carbon fiber reinforced plastics (CFRP) sold is below the amount of glass fiber reinforced plastics, their economic value is high. As carbon fiber production also

causes a high embodied energy, there is great incentive for recycling (Howarth *et al.*, 2014). Taking a gate-to-gate perspective Meng *et al.* (2018) assessed the primary energy demand and global warming potential (GWP) of different carbon fiber recycling techniques (mechanical, pyrolysis, fluidized bed, and chemical recycling process) as well as of conventional landfill and incineration. To account for changing material properties due to recycling activities the authors have considered two different carbon fiber replacement methods. The first is based on a variable fiber content. To compensate for the poorer mechanical properties of recycled carbon fiber relative to virgin carbon fiber, the fiber volume fraction of recycled carbon fiber reinforced plastic can be varied. The second replacement method is based on variable material thickness. The fiber content remains constant and component thickness is varied to account for differences in the mechanical properties (Meng *et al.*, 2018).

For primary energy demand as well as for GWP, carbon fiber recycling through pyrolysis, fluidized bed and chemical processes show significant advantages over conventional end-of-life treatments for both replacement methods. The advantages are thereby primarily based on the replacement of virgin carbon fiber by recycled carbon fiber (Meng *et al.*, 2018).

Fig. 5 illustrates the net environmental impact of recycling in terms of GWP using the case of pyrolysis recycling with the data of Meng *et al.* In addition to pyrolysis recycling itself, the process steps of transport and cutting must also be considered. The environmental impact indicator E_{repro} amounts for 2.9 kg CO₂eq/kg CFRP. The indicator for the landfill of an equivalent amount of CFRP equals $E_{\text{landfill}} = 0.14$ kg CO₂eq. Environmental benefits are only achieved by substituting virgin materials through recycled carbon fiber and byproducts such as oil/wax and gas. The authors assume that 0.88 kg virgin fiber production can be avoided with one kg CFRP waste assuming a variable fiber content. In total, a net environmental benefit of 23.6 kg CO₂eq/kg CFRP can be achieved with pyrolysis recycling. However, the authors emphasize that achieving the high level of environmental benefits from recycling also depends on the potential market (Meng *et al.*, 2018) (Fig. 6).

Conclusion

Composite materials continue to gain in importance and are used in various branches of industry. Due to their high stiffness and strength compared to low density, composites are particularly suitable for lightweighting and thus enable efficiency improvements during the use stage. Nevertheless, composite materials do not per se have a lower environmental impact than conventional engineering materials. In addition to the use stage, the other life stages such as materials production, part generation and the treatment at the end-of-life must also be taken into account. Life Cycle Engineering supports such a life cycle perspective and thereby helps to engineer products with the lowest possible environmental impact. The core methodology of LCE is life cycle assessment which aims to quantify the environmental impact of products and processes over their entire life cycle.

Due to an extension in application fields and more demanding requirements, the composite industry is experiencing a dynamic development. LCA-based LCE approaches need to be introduced to avoid problem shifting regarding potential environmental impacts of innovative composites. However, methodological challenges need to be addressed in this context. Two topics as well as potential challenges in the LCA-based LCE are exemplarily discussed in this article, namely the multifunctionality and the circularity of composite materials.

- Multifunctionality: with regard to multifunctionality, the aspect of allocation of environmental impacts presents a methodological challenge. Within the state of research, only one study could be identified that evaluated multifunctionality in the context of environmental impacts of composites, while other studies disregard the aspect.
- Circular Economy: with regard to circular economy and composite materials, the state of research currently focuses on recycling techniques. Since the various techniques differ in terms of process steps, material and energy consumption as well as emissions, an LCA-based engineering is essential to ensure that the recycling of composite materials does not have a negative impact on the environment. However, further research on higher-value reprocessing strategies is necessary in order to reuse products in a less material- and energy-intensive way. The extension of the life cycle, e.g., by means of appropriate maintenance strategies, should also be aimed for.

A last, but all the more important observation is related to the perspective of LCA-based LCE for composites. It can be concluded that most current efforts continue to focus on improving eco-efficiency rather than eco-effectiveness. However, it is essential that the limited carrying capacity of the planet is taken into account. In this context, increasing production volumes, due to a growing population and increasing affluence, as well as the associated overall environmental impacts, have to be considered. Thus, future research in LCE should focus on methods and tools that support the reduction of the overall environmental impact. The top-down perspective, as proposed within the Lyngby framework (see Section "Methodology"), can help reduce complexity and support the identification of the most relevant (foreground) systems.

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Carbon Footprint of Waste-Derived Composites

Ivan Deviatkin and Kaisa Grönman, Department of Sustainability Science, Lappeenranta–Lahti University of Technology LUT, Lappeenranta, Finland

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Glossary

Carbon footprint (CF) (or a carbon footprint of a product) The sum of greenhouse gas emissions and greenhouse gas removals in a product system expressed as carbon dioxide equivalents ($\text{CO}_{2\text{eq}}$) and based on a life cycle assessment using a single impact category of climate change.

Elementary flow Material or energy entering the system being studied that has been drawn from the environment without previous human transformation, or material or energy leaving the system being studied that is released into the environment without subsequent human transformation.

End-of-life (EoL) A life cycle stage of a product indicating that the product under study is ready for disposal, recycling, reuse for different purposes, or energy recovery.

Greenhouse gas A gaseous constituent of the atmosphere, both natural and anthropogenic, that absorbs and emits radiation at specific wavelengths within the spectrum of infrared radiation emitted by the Earth's surface, the atmosphere, and clouds.

Life cycle assessment (LCA) Compilation and evaluation of the inputs, outputs, and potential environmental impacts of a product system throughout its life cycle.

Waste-derived composites (WDC) Composite materials which are manufactured fully or to a very large extent from waste feedstock.

Introduction

During an age of rapid population growth (mostly in developing countries) and increasing living standards (mostly in developed countries), environmental conservation has become a cornerstone of the long-term survival of humankind. Deforestation, eutrophication, acidification, toxic impacts, drought, wildfires, accelerated extinction of species, and climate change are all threatening the fragile stability of natural ecosystems, as well as the well-being of modern humans.

Increasing mobility, mechanization, automation, and industrialization have all made us dependant on fossil fuels, which have been used as a primary source of energy around the globe. Long before we were alerted to climate change, fossil fuels were incinerated in large amounts, releasing substantial amounts of fossil-based carbon dioxide. The production of many materials, such as different types of synthetic polymers, requires fossil fuel consumption, either as a feedstock for the materials or as an energy carrier.

One effective pathway towards the reduction of human environmental impacts is the use of already manufactured materials and products to satisfy the existing needs of society. This implementation represents one of the pillars of the circular economy, focusing on the reuse, recycling, and recovery of materials within technical and biological cycles. Here, waste of various origins is seen as a feedstock for numerous production processes.

The highest recycling efficiency can be reached either through closed-loop recycling, whereby materials are circulated within the same manufacturing process, or open-loop recycling, whereby no changes occur in their inherent properties. However, closed-loop recycling often requires a separate collection system of waste, such as those of aluminum cans or PET bottles implemented via take-back systems, and is thus hard to set up. Instead, open-loop recycling can be proposed to ensure the utilization of materials in other production processes which could use waste streams with higher levels of impurities and less sensitivity to the homogeneity of feedstock.

Composites are a unique materials niche which allows for the utilization of various feedstock. Furthermore, polymer composites are renowned for their suitability for utilizing a wide range of waste, including recycled plastic and wood waste, among other waste fractions. Specific properties of WDC are then tailored using coupling agents, processing aids, stabilizers, flame-retardants, antioxidants, ultra-violet stabilizers, and antimicrobial aids. Finally, WDC are oftentimes used in technical applications, thus allowing for the possible presence of impurities therein.

The production of WDC has the potential to reduce environmental impacts, including mitigation of climate change. This could be achieved through the avoidance of feedstock production, which usually has a higher impact than the impacts associated with the collection of waste and its possible pre-treatment, such as sorting, drying, or size reduction. Furthermore, WDC can be recycled back into composites, thus extending the lifetime of the materials embedded in them. Finally, when WDC are manufactured, they could replace certain products on the market, such as those made of primary plastic, wood, or even metals.

However, the CFs of composites differ greatly. The major contributing factors are the manufacturing processes used, the amount of additives needed, the carbon intensity of the grid, their recyclability, and the EoL method. In some cases, the CF of WDC can exceed that of the baseline solution without their production.

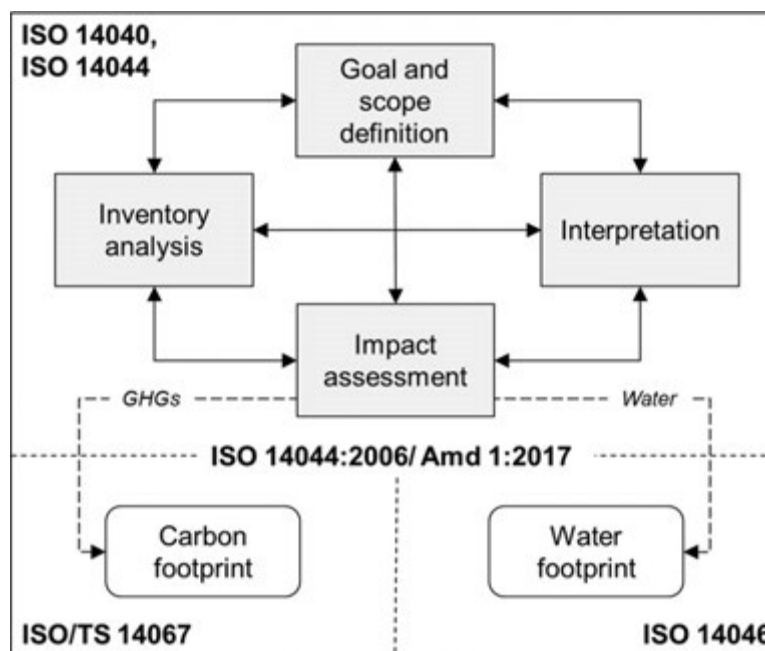


Fig. 1 Phases of LCA and their relation to carbon and water footprints.

This article of the Encyclopedia offers insights into the assessment of the CF of composites produced from waste. It will introduce the core principles of LCA and its specific derivative, the CF. The concept of carbon handprint will also be spotlighted. The article will provide the principles of GHG emissions and removals accounting. Finally, it will elaborate on various possibilities of climate change impact from WDC.

LCA Methodology

LCA methodology is governed by Standards ISO 14040 on the principles and framework of LCA (SFS-EN ISO 14040, 2006), as well as ISO 14044 on requirements and guidelines (SFS-EN ISO 14044, 2006) which was amended in 2018, mostly as concerns the critical review and footprint studies. The ISO standards, however, leave LCA practitioners with a range of methodological choices. Therefore, additional literature also exists to guide them, such as the International Reference Life Cycle Data System (ILCD) handbook (Joint Research Center – Institute for Environment and Sustainability, 2010a,b) and the Product Environmental Footprint (PEF) (Zampori and Pant, 2019), among others. Also, industry- or activity-specific LCA guidelines exist, such as those by the World Steel Association (2011) or International Energy Agency (2011).

Each of the LCA studies essentially follows the following phases: (a) goal and scope definition, (b) inventory analysis, (c) impact assessment, and (d) interpretation. Despite having a specific order, these phases are not consecutively arranged in LCA, owing to its iterative nature; thus, choices made during the goal definition could be adjusted based on the inventory or impact assessment stages. Each of the phases is important for the overall quality of the LCA studies and should not be omitted.

When LCA studies focus on specific areas of concern, they are termed footprint studies. Footprint studies can have their own methodology, such as those for water footprint calculations (SFS-EN ISO 14046, 2016) and CF calculations (SFS-EN ISO 14067, 2018). Footprint studies delve deeper into aspects of impact assessment in a specific area of environmental concern, such as climate change, which are otherwise not covered comprehensively in the overarching LCA standards ISO 14040/44. Fig. 1 shows the key phases of LCA, as well as the connection of footprint standards with such overarching standards.

CF Methodology

CF studies should follow Standard (SFS-EN ISO 14046, 2016) to ensure the correctness of calculations and be suitable for external communication. According to the Standard, a CF is the sum of “GHG emissions and GHG removals in a product system expressed as CO₂ equivalents and based on a life cycle assessment using a single impact category of climate change.” Therefore, to proceed with a CF study, each practitioner is expected to know LCA methodology.

An assessment of the climate change impacts of composite materials within the CF methodology is expected to be based on a declared unit as opposed to the functional unit, owing to the limitation of its life cycle in terms of the life-cycle stages included. When such studies include only cradle-to-gate impacts, they are called partial CFs (PCFs) of products. This is explained by the fact

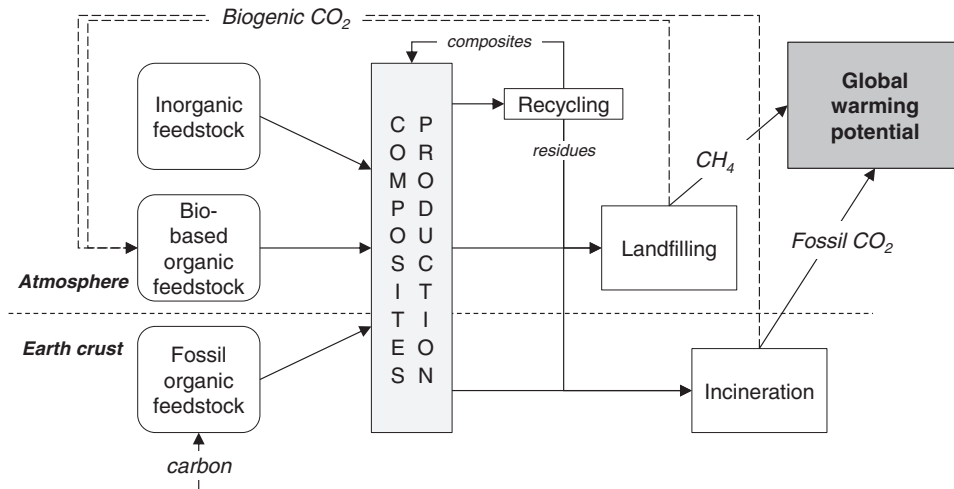


Fig. 2 Implications of various waste feedstock on the climate change. The use phase is omitted since it has no difference between the various feedstock.

that the composites could be used later in various applications, thus having different impacts before their EoL is reached. The declared unit is oftentimes related to a specific amount of the studied product. In the case of composites, this would be a mass-based unit of, for example, 1 kg of composites or a volumetric unit of, for example, 1 m³ of composites.

Carbon Handprint Approach

The carbon handprint approach was developed to quantify the positive climate impacts of products. In contrast to the CF, the carbon handprint presents the amount of reduced greenhouse gas emissions when a product is being used by a certain customer. The carbon handprint is always calculated against a fairly set baseline. A CF of a baseline solution and a CF of the alternative, often innovative, solution are quantified, and their difference equals to the carbon handprint. The carbon handprint approach can be utilized alongside the footprints to further communicate the positive impact of the products. Besides, the handprint approach may reveal product development needs, if the baseline solution already has a lower footprint (Pajula *et al.*, 2018; Grönman *et al.*, 2019).

In the carbon handprint assessment, it is important to recognize the *possible contributing mechanism*, which can allow footprint reduction (Pajula *et al.*, 2018; Grönman *et al.*, 2019). In the case of WDC, several benefiting factors may contribute to the carbon handprint:

- *Less GHG-intensive material use:* Depending on the intended application, WDC material or product may replace non-renewable materials, that might be of virgin origins and GHG-intensive material. When waste is utilized as raw material, material-use efficiency may be improved, given that processing does not require large additional material inputs.
- *Less GHG-intensive energy use:* Production of WDC can usually be assumed to be less energy-intensive than producing the material from primary origins, due to the lower energy required to melt smaller amounts of plastic.
- *Increased lifetime and performance:* When using WDC as raw material for a product, one must be certain that the properties of the composite material are comparable to, or better than, the properties of the material from which the product would otherwise be manufactured. The lifetime of the product should not be allowed to decrease even if WDC are utilized as a raw material. However, in this case, the lifetime of the (waste) material is prolonged when it reaches a second life as a composite material.
- *Reduced waste and losses:* Using waste as a raw material in the composite production naturally falls into the category of possible carbon handprint contributors. However, the producer of the WDC should acknowledge that the impurities in the waste feedstock, such as unintended waste fractions, may cause losses in WDC production.
- *Increased carbon capture and storage:* Utilizing waste as a raw material rather than primary sources may contribute to carbon sequestration if land use change can be avoided. Besides, the carbon already stored in the composite material remains unrealized for a longer period.

Inventory and Interpretation of GHG Emissions and Removals

The CF method manifestly accounts separately for biogenic and fossil GHG emissions and removals occurring in the studied system, as well as those occurring as a result of direct land use, or as aircraft emissions. Additionally, climate change impacts from

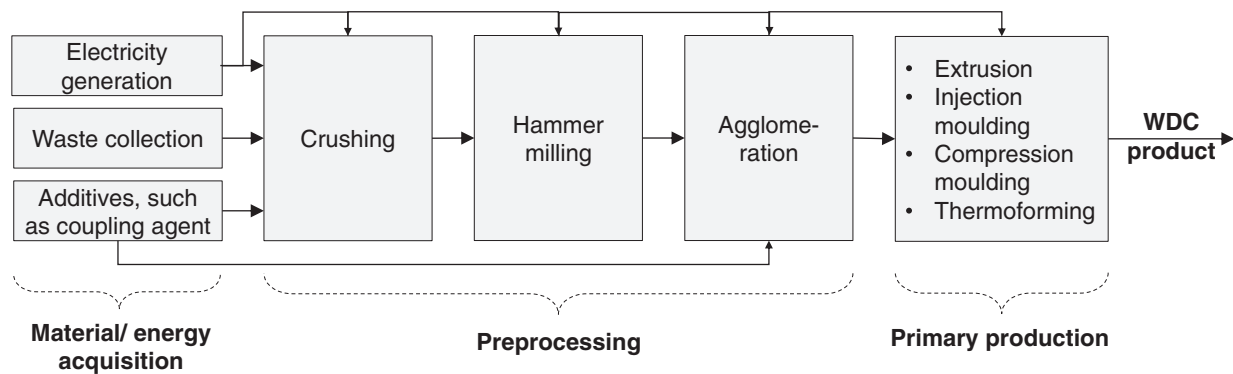


Fig. 3 Generalized production process of waste-derived composites.

indirect land use change and land use can be included. The sequestration of carbon dioxide in biomass shall be characterized as $-1 \text{ kg CO}_{2\text{eq}}$ per 1 kg CO_2 when entering the product system and as $+1 \text{ kg CO}_{2\text{eq}}$ per 1 kg CO_2 when being emitted back into the environment as carbon dioxide, thus resulting in zero net climate change impacts. The exception to the rule holds true when the carbon is not being oxidized to carbon dioxide but is converted to methane, non-methane volatile organic compounds, and other precursors, which is the case for organic matter used for landfill. To assess other GHGs and their characterization factors, the latest IPPC reports should be consulted.

Waste-Derived Feedstock and its Climate Change Implications

From the CF study point of view, three major types of waste-derived feedstock can be distinguished: inorganic feedstock, bio-based organic feedstock, and fossil organic feedstock. The differences in feedstock can be accounted for in CF studies through their avoided environmental impacts if having a consequential approach to the study. Furthermore, the content of different feedstock will imply variations in the EoL impacts of WDC. The implications of the various feedstock for climate change impacts are illustrated in [Fig. 2](#).

Inorganic feedstock, such as mineral wool or plasterboard, is mostly used as a filler. Its inorganic nature implies potentially low impacts on climate change due to its conventional EoL, which could be landfill or re-use in the construction industry. Therefore, such material is not expected to contribute to a significant reduction of climate change impacts when used in composites and not disposed of conventionally. At the EoL, inorganic materials do not generate emissions during incineration or contribute to the generation of landfill gases, if used as landfill.

Bio-based organic feedstock, such as agricultural residues, waste wood, and primary sludge, is also mostly used as a filler. Bio-based organic feedstock contains biogenic carbon, which is a part of the natural carbon cycle being sequestered during biomass growth. When bio-based organic feedstock is incinerated, the emissions are accounted for as climate-neutral. However, the same feedstock placed in landfill results in the formation of landfill gas containing methane, which contributes to climate change. It is worth mentioning that methane has a 28 times stronger radiative forcing than carbon dioxide over a time horizon of 100 years; that is, GWP_{100} of methane is $28 \text{ kg CO}_2\text{-eq}$ (IPCC, 2014).

Fossil organic feedstock, such as various plastics, is used as a matrix material. Such feedstock contains carbon, which contributes to the global warming potential when released into the atmosphere. The incineration of fossil organic feedstock has the worst climate change impacts since all the carbon is converted into fossil-based carbon dioxide. Using such feedstock as landfill has a lower impact on climate change. However, this disposal method leads to increased abiotic resource depletion since more fossil resources would be required to replace that disposed of in landfill. An overview of the impacts from various feedstock is presented in the study by [Sormunen et al. \(2021\)](#).

Climate Change Impacts of WDC

Production Process

The environmental impacts of WDC start with the collection of waste from its place of generation ([Fig. 3](#)). The waste can originate from various activities, such as industrial production or construction, or can be commercial, demolition, or agricultural waste. When waste is collected for composite production, it first has to be pre-processed. Crushing and hammer milling are used to reduce the particle size, whereas magnetic separation might be used to remove undesired metal parts, such as nails ([Liikanen et al., 2019](#)). Agglomeration is used to blend the pre-processed waste feedstock and additives into compounds. The primary composite production process is similar to the polymer production process, the most common processes being extrusion, injection molding, compression molding, or thermoforming ([Gardner, Han and Wang, 2015](#)).

Table 1 Carbon footprint of certain raw materials and energy sources

Activity	CF, CO _{2eq} unit ⁻¹	Reference
Transportation, incl. diesel production, tkm	88 g	(LIPASTO, 2019; PE International, 2014)
Electricity, kWh	74 g Brazil, 136 g Finland, 453 g USA, 555 g China	(Carbon Footprint Ltd, 2020)
Plastic granulates production, kg	1.80 kg HDPE, 1.87 kg LDPE, 2.19 kg PET	(PlasticsEurope, 2020)
Dried timber, kg	0.15 kg, (– 1.5 kg embodied)	(RTS EPD Ympäristöseloste, 2019)

In the extrusion process, continuous linear profiles are formed as melted thermoplastic is forced through a die (Migneault *et al.*, 2009). In injection molding, the heated mixture is fed into a highly pressurized mold cavity of the desired shape. In compression molding, a pre-shaped sheet is placed into a mold that is then forcefully enclosed under high pressure, whereas in thermoforming, the sheet is first heated and then formed into the desired shape with a mold.

Impact of Production

The CF of composite production can be estimated based on the consumption of raw materials and energy in the production process. Table 1 summarizes the CFs of the transportation process, electricity generation, and plastic and wood production. Relying on the data presented in Table 1 and the life-cycle inventory presented in the study by Liikanen *et al.* (2019), a PCF can be estimated for a declared unit of 1000 kg (or 1 metric ton) of WDC. The production recipe for WDC in the study by Liikanen *et al.* (2019) comprises 54% wood waste, 40% plastic waste, and 6% additives. Since WDC production can utilize waste, its production efficiency is close to 100%; hence, to produce 1000 kg of WDC, the same amount of feedstock, such as fillers, matrix, and additives, would be needed.

Transportation distances for waste collection can range significantly from one place to another. If the waste feedstock is collected from a distance of 100–200 km, the CF of the collection would be 8–16 kg CO_{2eq} per 940 kg. The additives might be transported from a more remote location using different transportation modes, such as ocean-going container ships. However, the impact is not expected to be high due to the relatively small amount of additives needed and the expected low impact of ocean-going transportation modes. Regarding the impact of additive production, manufacturing 30 kg of maleated polypropylene and 30 kg of a processing aid has a CF of around 100 kg CO_{2eq}. The impact of the additives might vary if other materials are used or the amounts are different. The electricity consumption for pneumatic moving, crushing, hammermilling, agglomerating, and extruding is around 1.6 MWh per 1000 kg of WDC. If injection molding is used, then the electricity consumption would increase to 2.3 MWh per 1000 kg of WDC. Therefore, the CF of electricity provision to the WDC production process would range between 210 and 310 kg CO_{2eq} in the case of Finland and 870 and 1270 kg CO_{2eq} in China.

The waste used in the WDC production process is commonly not considered to have any environmental impacts from the previous life cycles of the materials under the so-called “zero-burden” approach (Ekvall *et al.*, 2007). However, the zero-burden approach is being questioned under the upcoming implementation of a circular economy in which all waste is supposed to be raw material in another production process (Djuric Ilic *et al.*, 2018). In such a case, allocation should be applied following the most recent LCA guidelines, for example those developed for paper products in Europe (Hohenthal *et al.*, 2019). Summing all the impacts from transportation, production of additives, and electricity generation, the cumulative impact from WDC production totals 310–420 kg CO_{2eq} in Finland and 970–1380 kg CO_{2eq} in China. The PCF of WDC production is lower than that of HDPE production, of 1800 kg CO_{2eq}. The PCF of wood, of 150 kg CO_{2eq} per 1000 kg of wood, is, however, considerably lower than that of WDC.

Impact of Use

When WDC are used, they would usually be considered as a replacement for a specific product made of alternative materials. The replacement of plastics has a higher potential for the reduction of environmental impacts due to its higher CF (Table 1). However, in the case of wood, WDC production could also be beneficial owing to its specific properties, such as moisture and UV resistance, higher strength, lack of treatment needs, and production of complex geometrical forms. When considering the application of WDC in the production of wooden pallets, substantial benefits could be expected from the better durability of pallets made of WDC, assuming their performance is closer to that of plastic, rather than wooden, ones (Deviatkin and Horttanainen, 2020). However,

use phase impacts are always case-specific, so making a PCF of WDC is beneficial to ensure the use of such data by LCA practitioners working on attributional or comparative LCAs of products made of WDC.

End-of-Life Impacts

WDC can have several EoL possibilities, of which recycling is the most circular. WDC are suitable for closed-loop recycling, for which only additives, such as a coupling agent, would be needed to ensure their technical properties meet specific requirements. In this case, the CF would be formed similarly as when producing WDC from waste, that is, transportation of WDC waste to the WDC production factory, followed by crushing, milling, agglomeration, and extrusion of molding. Here, the benefits of recycling would relate more to the extended lifetime of the material than the avoided impact from raw materials acquisition, which is the case for the recycling of paper, plastic, or metals, when materials produced from fossil resources are being replaced. In the case of WDC recycling, only zero-burden waste is replaced.

If closed-loop recycling cannot be implemented, which may well be the case since it usually requires the development of the take-back infrastructure enabling products made of WDC to be returned to the factory, their incineration with energy recovery can be practiced as a disposal option following recycling in the European waste hierarchy ([European Parliament and the Council of the EU, 2008](#)). In this case, the WDC are sent for thermal treatment, whereby electricity and thermal energy is generated. Energy outputs can, thus, replace the same amount of energy produced in a specific country. When considering electricity and heat substitution, a consequential approach is recommended ([Ekvall and Weidema, 2004](#)). In the consequential approach, additional electricity and thermal energy provision are assumed to be compared with the replacement of the electricity and thermal energy sources otherwise being phased out in the energy system of a specific country or a region. Such sources are usually coal, natural gas, and crude oil-based fuels.

Incineration of wood has almost no impact on climate change if carbon is oxidized completely and no other GHG emissions are being formed, such as methane or nitrous oxides, when providing electricity. Neither does the incineration of WDC containing inorganic materials, such as mineral wool, contribute to climate change, but their presence in the incineration process does not generate any electricity; on the contrary, it reduces the total energy efficiency due to energy consumption in the heating process. Finally, plastics have a high heating value and are highly calorific fuels. However, due to their fossil origin, plastic incineration has a tremendous impact on climate change. Plastics generally contain 80% carbon, so the incineration of 1 kg of plastics would result in emissions of 2.9 kg CO_{2eq}, stoichiometrically calculated from the mass of carbon oxidized to carbon dioxide.

Finally, WDC can also be used as landfill if legislation allows. In Finland, landfilling is no longer possible for wood and plastic waste due to the ban on organic waste landfill that has been in force since 2016. However, in many countries, landfilling is still widely practiced, posing the potential situation of plastics and composites outperforming wood. The reason for this is the stability of plastics and WDC to biological decay, whereas wood is subject to biological decay during landfill. Landfilling 1 kg of wood could emit 1.3 kg CO_{2eq}, of which some 95% is contributed by the methane formed. It should be noted that there is a large variation between existing landfill types so the actual CF from wood landfill could range significantly, depending on the implementation of the landfill gas collection system, its efficiency, the type and location of the landfill, and so on.

Conclusions

Composites represent a promising solution for the achievement of circular economy principles which are being widely developed. The circular economy can prosper from the utilization of waste in the production of composites, or the so-called WDC. WDC can be manufactured from a wide range of feedstock, such as bio-based organic materials such as wood waste and straw; mineral fillers such as mineral wool; and fossil-based organic materials, such as various plastics, which act as a matrix. The properties of WDC can be adjusted based on the application, using a wide range of additives, including coupling agents, lubricants, UV resisting agents, colorants, antimicrobial aid, and so on. WDC can be produced through several methods, such as extrusion to produce continuous linear profiles, and various types of molding and thermoforming allowing products of the desired shape to be manufactured. This wide variability in the possible feedstock, properties, and applications makes WDC a unique material.

However, the impacts of WDC on climate change are not always straightforward and positive since they always depend on the operating environment. If WDC are produced to potentially replace wood with a short service life, then the climate change impact of their production and consequent incineration at the EoL is expected to be higher than that of wood. However, if WDC are produced as a unique material serving purposes potentially not achievable with wood, due to their strength or geometrical shape, but which can still be achieved with plastic, then the WDC could outperform plastics due to their lower production and EoL impact. However, the production of WDC itself could also be important if it takes place in a country where the CF of electricity is high because an absolute majority of the CFs of WDC originate from electricity consumption.

Variability of the feedstock, products which may be made from WDC, and EoL options together make it practically impossible to draw any specific conclusion as to their superiority over alternative products or on their own. Each case must be assessed according to specific conditions and primary data on the production of the composites, which is still widely lacking as few studies have been conducted on this subject. The lack of verified primary inventory data on WDC production is pointed out and should be addressed in future research.

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Wimao.

Energy Management for Composite Materials Manufacturing: Energy and Exergy Analyses

Lorna Fitzsimons, Advanced Processing Technology Research Centre, School of Mechanical and Manufacturing Engineering and the Water Institute, Dublin City University, Dublin, Ireland

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Nomenclature

ASHRAE American Society of Heating and Air-Conditioning Engineers

BAAM Big area additive manufacturing

CVD Chemical vapor deposition

DMD Directed material/energy deposition

EDM Electrical discharge machining

EnPIs Energy performance indicators

FDM Fused deposition modeling

IEA International Energy Agency

Symbols

$\text{gCO}_2 \text{ MJ}^{-1}$ Grammes of CO_2 per megajoule

e Specific exergy (J kg^{-1})

$\dot{E}_D$ Rate of exergy destruction (W)

g Gravitational acceleration (m s^{-2})

h Specific enthalpy (J kg^{-1})

h_0 Specific enthalpy at the defined dead state (J kg^{-1})

ktoe Kilotonnes of oil equivalent

$\dot{m}_i$ Mass flow rate of flow i (kg s^{-1})

$\dot{Q}_i$ Rates of heat transfers at various locations i on the system boundary (W)

s Specific entropy ($\text{J kg}^{-1} \text{ K}^{-1}$)

s_0 Specific entropy at the defined dead state ($\text{J kg}^{-1} \text{ K}^{-1}$)

T_i Instantaneous temperature on the system boundary (K)

T_0 Dead state temperature (K)

V Velocity at thermodynamic state (m s^{-1})

V_0 Velocity at the dead state (m s^{-1})

$\dot{W}_i$ Rates of exergy transfer associated with work transfer i (W)

z Elevation at the thermodynamic state (m)

z_0 Elevation at the dead state (m)

Introduction

Effective energy management is important to reduce operating costs, but also to reduce the impact that manufacturing activities have on the environment. According to 2018 data, industrial energy consumption accounted for approximately 29% of global energy consumption (International Energy Agency, 2021). In the United States, industry is the largest end-user of energy, with the manufacturing sector in particular responsible for 70% of industrial energy usage; this is followed by the mining sector at 20% (U.S. Department of Energy, 2018). At the global level, there is a relatively even split between the travel and industry sectors, which combined, made up 58% of global energy use in 2018 (International Energy Agency, 2021). High energy consumption can be a significant operating expense for many sectors, and is often an important contributor to an organization's carbon footprint. For manufacturing processes in general, the most common forms of energy usage are electrical and thermal energy. Electrical energy is used for powering processing equipment and lighting, and thermal energy is commonly in the form of fossil fuels (oil/gas) that are used in boilers and gas turbines. Energy usage in manufacturing facilities can be split into the direct energy requirements of the processing equipment, and the indirect energy usage and losses associated with the building fabric, and maintaining the necessary environmental conditions for both process and staff requirements. This plant/processing breakdown is somewhat analogous to the economic concepts of direct and indirect costs, and fixed and variable costs. In complex, clean manufacturing environments, the energy requirements to maintain cleanroom environmental conditions can be significant. Generally, cleanrooms require strict temperature and humidity controls, frequent volumetric air changes, and maintaining a positive pressure relative to areas in direct proximity to the cleanroom.

Global manufacturing energy consumption impacts on the environment throughout the multiple phases of the energy life-cycle. The magnitude of this contribution depends on the quantity and type of energy consumed, and is also a function of the primary energy mix that is used to produce the electricity in a given location. With regard to fossil fuel based electricity and thermal energy, these phases include: the extraction and refining of primary energy resources; the production of electricity in power plants; the transmission of electricity; the transport of gaseous, liquid and solid fossil fuels; and the direct emissions produced from on-site thermal energy systems. Each of these phases has inherent inefficiencies that result in cumulative thermodynamic losses. The main environmental impact category of fossil-fuel based electricity production is global warming potential; however, the presence of SO_2 and NO_x also means that there are acidification and eutrophication impacts (Turconi *et al.*, 2013).

As mentioned, emissions can be categorized into direct and indirect emissions. The U.K. Government's greenhouse gas conversion factor document further breaks down emissions into three scopes: Scope 1 are direct emissions, i.e. emissions that are controlled by the organization (boilers, furnaces, vehicles); Scope 2 are indirect emissions that results from an organization's energy use, but are outside your control (purchased electricity, heating/cooling); Scope 3 are other indirect emissions such as business airline travel (U.K. Government, 2021).

Composite materials refer to a combination of two or more different materials that meet the following criteria: the materials are chemically distinct; their phases are insoluble; and they have a recognizable interface (Kalpakjian and Schmid, 2009). Composites therefore encompass a wide range of materials and applications; they can be generally categorized into metal-matrix, polymer-matrix

and ceramic matrix composites. Importantly, composites are specifically engineered and functionalized to take advantage of their synergistic and superior engineering properties. The applications for composites are ubiquitous: aerospace, large structural components, electronics, sports goods etc. Because of the wide range of composite materials and their far-reaching applications, there are many manufacturing processes that are used in their production. Common composite manufacturing processes for polymer-matrix composites include variations of the molding, extrusion and casting processes, for both thermosets and thermoplastics. However, there are frequently additional processing steps required to integrate the reinforcement materials to ensure adequate bonding, e.g., impregnation, which can increase processing steps, cycle time and cost. Metal-matrix composites are manufactured using liquid phase, e.g., casting processes that include both the metal-matrix and the reinforcement material; solid-phase processes that utilize powder-metallurgy techniques; and two-phase processes such as rheocasting or spray atomization and deposition. With regard to ceramic-matrix composites, common manufacturing processes include infiltration processes (e.g., slurry and chemical vapor), sintering and chemical synthesis processes. In addition to the more common manufacturing processes, other additive manufacturing techniques are becoming prevalent, processes such as fused-deposition modeling, 3D printing, selective laser melting/selective laser sintering. An in-depth presentation of all of these processes is found in (Kalpakjian and Schmid, 2009), and later editions.

Energy is a key manufacturing input. Electrical and thermal energy are required for high temperature phase changes, to power processing equipment, and for the associated manufacturing and business facilities. In terms of the manufacturing processes, it is evident that the energy requirements will differ according to the specific process under consideration, which is itself a function of the composite material, but also the type and application of the manufacturing process. For example, metal-matrix composites have higher melting temperatures than polymers, and composites produced and used in the highly controlled manufacturing environments, typical of the semiconductor or biomedical industries, will inherently have a higher energy overhead than those of less critical applications.

This article is broken down into several sections. First, the relationship between energy and climate change is discussed. A brief introduction to some relevant aspects of energy theory is presented; which includes a discussion of power factor and commonly used devices like motors. Exergy analysis is introduced, and the energy intensity of several manufacturing processes in terms of both exergy and energy intensity is reviewed. Finally, the concept of energy management is introduced and discussed in relation to the energy hierarchy, energy management systems, energy auditing, and energy benchmarking.

Energy and Climate Change in Context

The sustainability of composite materials is a broad topic that incorporates all aspects of the composites product life cycle: the extraction of fossil fuels; the production of plastics/metals/ceramics; the manufacture of composites; the use of composites; their end of life, whether that be reuse, recycling, energy recovery, or landfill. Energy is just one consideration in this broader context. However, it is an important factor in the various phases of the product life cycle of composite materials, a concept that is captured and quantified in the embodied energy of materials. The main focus of this article, however, is energy and specifically energy management in composites manufacturing.

The Kyoto protocol was adopted in 1997, whereby the world's industrial nations recognized the threat of climate change, and agreed to mitigate greenhouse gas emissions. It is widely accepted that anthropogenic activities have a major impact on greenhouse gas emissions, and consequently global warming and climate change. Global energy consumption is one of the key contributors to greenhouse gas emissions, and in particular the combustion of fossil fuels. It is therefore interesting to track how global energy supply and consumption of fossil fuel based energy streams has changed since Kyoto, in both absolute and relative terms. First, primary energy supply is assessed; second, total final energy consumption is considered in general terms, and then at the sectoral level.

According to data from the International Energy Agency (IEA), from 1997 to 2018, the supply of oil has increased by 26.3%, natural gas by almost 71%, and perhaps more alarmingly, coal by almost 73%; see **Table 1** (International Energy Agency, 2021).

The total final energy consumption by source follows a similar trend, with significant increases in final energy consumption for all energy sources, especially coal and electricity (**Table 2**).

Again using IEA data (International Energy Agency, 2021), the total final energy consumption breakdown by sector is shown in **Table 3**, in terms of magnitude, and in **Fig. 1**, in terms of relative consumption.

At the sectoral level, it is evident from **Fig. 1** that the industry and transport sectors account for the majority of final energy consumption. Their combined final energy demand represented 54% of total final energy consumption in 1998, and this share increased to 58% in 2018. One other main change of note was that the final energy demand of the residential sector reduced from 26% to 21%.

Table 1 Energy supply data

	1998 (ktoe)	2018 (ktoe)	Absolute increase (ktoe)	% increase (%)
Oil	3,560,865	4,496,998	936,133	26.3
Natural gas	1,908,341	3,261,595	1,353,254	70.9
Coal	2,220,003	3,838,326	1,618,323	72.9

Note: Adapted from International Energy Agency, 2021. [Online] Available at: <https://www.iea.org/data-and-statistics?country=WORLD&fuel=Energy%20supply&indicator=TPESbySource> (Accessed 25.01.2021).

Table 2 Total final energy consumption data by energy source

	1998	2018	Absolute increase (ktoe)	% Increase (%)
Oil	2,962,185	4,038,502	1,076,317	36
Natural gas	1,041,750	1,611,345	569,595	55
Coal	588,634	994,497	405,863	69
Electricity	1,013,198	1,918,779	905,581	89

Note: Adapted from International Energy Agency, 2021. [Online] Available at: <https://www.iea.org/data-and-statistics?country=WORLD&fuel=Energy%20supply&indicator=TPESbySource> (Accessed 25.01.2021).

Table 3 Total final energy consumption by sector

Sector	1998 (ktoe)	2018 (ktoe)
Industry	1,788,057	2,839,313
Transport	1,852,005	2,890,900
Residential	1,744,114	2,109,205
Commercial and public services	534,410	808,619
Agriculture/Forestry	171,701	214,719
Fishing	5788	7005
Non-specified	76,014	151,179
Non-energy use	571,037	916,762

Note: Adapted from International Energy Agency, 2021. [Online] Available at: <https://www.iea.org/data-and-statistics?country=WORLD&fuel=Energy%20supply&indicator=TPESbySource> (Accessed 25.01.2021).

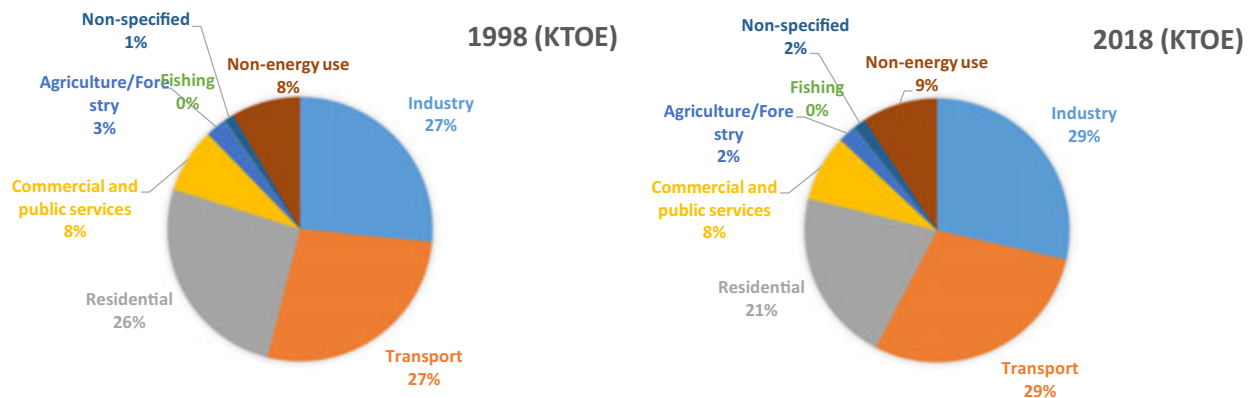


Fig. 1 Total final energy consumption by sector, 1998 and 2018, adapted from IEA data. Reproduced from International Energy Agency, 2021. [Online] Available at: <https://www.iea.org/data-and-statistics?country=WORLD&fuel=Energy%20supply&indicator=TPESbySource> (Accessed 25.01.2021).

In addition, the carbon intensity of industrial energy consumption, measured in grammes of CO₂ per megajoule of energy (gCO₂ MJ⁻¹), also increased, albeit slightly, from 51.5 gCO₂ MJ⁻¹ in 1998–51.8 g CO₂ MJ⁻¹ in 2018. Taking these factors into account, it is clear that not only has global energy supply and consumption increased in absolute and relative terms, the carbon intensity of that energy consumption has also increased. Given that one kilotonne of oil equivalent (ktoe) is approximately equal to 4.187×10^7 MJ, the increase in industrial energy consumption from 1998 to 2018 was responsible for an additional 1.32×10^{13} gCO₂. Energy consumption can be reported using multiple metrics e.g. per capital, per unit GDP. However, in terms of climate change, metrics are meaningless; it is absolute quantities that count. That said, on both the global and sectoral scale, it is difficult to reconcile the early ambition of the Kyoto protocol with the actual primary energy supply and final energy consumption data in the intervening period.

Energy Hierarchy

Managing energy resources effectively means different things to different stakeholders. It can be interpreted as increasing energy efficiency, increasing the number of renewables and thus reducing the dependence on fossil fuels, and reducing energy

consumption in absolute terms. All of these options are viable; however, they do not all have the same impact in terms of sustainability. The energy hierarchy was developed to rank various options from the least to the most sustainable, see Fig. 1 (Institute of Mechanical Engineers, 2020). It is evident that energy demand reduction is the preferred option. Energy has its own life cycle and inherent losses, i.e. the energy losses that occur from the extraction of fossil fuels, the production, and transmission/transportation to end-use. If the demand for energy can be reduced, these cumulative losses along the length of the energy pipeline are negated. The term *negawatt* was coined by Lovins in the 1980s to communicate the idea of saved cumulative energy by means of energy demand reduction.

Second in the hierarchy is energy efficiency, i.e., doing more with what the energy that is currently used, or doing the same with less. From the manufacturing energy management perspective this is a very effective approach. For example, in simple terms, if the energy efficiency of a motor is improved, the motor requires less electrical energy, resulting in lower energy costs for the manufacturing plant. Similarly, in a well-insulated building, if the building fabric heat losses are reduced less thermal energy is required. However, in broader terms, the rebound effect in Jevons paradox suggests that increasing energy efficiency can lead to undesirable outcomes. With regard to environmental economics, the Jevons paradox suggests that energy efficiency results in a positive feedback loop: relative cost is reduced, which in turn increases demand, ultimately resulting in higher energy consumption in absolute terms. There is debate in the literature about various aspects of Jevons' paradox, particularly the difficulty of empirically testing the rebound effect (Sorrell, 2009). However, Sorrell does also suggest that the rebound effects may in fact be larger than previously thought, and that the part energy plays in driving economic growth is underestimated, thereby suggesting that rebound effects potentially have "profound implications for energy and climate policy (Sorrell, 2009)".

The third tier in the energy hierarchy concerns the use of renewable and sustainable resources, for example, hydroelectric power, and wind energy. Essentially these technologies harness the climate in terms of rainfall, snowfall and wind to produce electrical energy. As such they do not involve the extraction and consumption of fossil fuels, so in comparison with oil, coal and natural gas they are considered to be a far more sustainable resource. Sustainability, in the longer term, of course assumes that climate patterns will not change in the geographic region where the hydroelectric and windfarm are deployed. In addition, there are shorter-term sustainability considerations. For windfarms, the life-cycle assessment of wind turbines is crucial in terms of their manufacturing materials, and their maintenance and decommissioning. With regard to hydroelectricity, the benefits are significant with respect to carbon mitigation and flood prevention; however, there remain significant social and ecological issues associated with building and operating dams. The Three Gorges dam is perhaps now infamous, and clearly illustrates the inherent trade-offs between often competing environmental impacts. On the one hand, the Chinese dam mitigates greenhouse gas emissions in comparison to fossil fuel power plants and provides flood protection. On the other hand, however, the Three Gorges dam has resulted in catastrophic social and environmental consequences including population displacement, landslides, increased pollution and risk of species extinction (Stone, 2011).

Tier 4 in the energy hierarchy concerns technologies such as carbon capture and sequestration, nuclear power, and using substituting technologies such as hydrogen for carbon based fossil fuels such as natural gas (Institute of Mechanical Engineers, 2020). The Institute of Mechanical Engineers make a case why these technologies are less than ideal: power plant efficiency issues with carbon capture and storage and inefficiencies associated with current hydrogen production technologies (Institute of Mechanical Engineers, 2020). There has been ongoing debate about the advantages and disadvantages of nuclear for some time now. The least sustainable scenario is business as usual (Fig. 2).

The energy hierarchy is analogous to the inverted triangle, waste hierarchy model, which was developed as part of the EU Waste Framework Directive (Directive 2008/98/EC). In a similar manner, the main focus of the waste hierarchy, in terms of the impact on sustainability, should be avoiding waste in the first place, with disposal being the least favorable option.

Energy Theory

Energy theory is both broad and deep. It is not possible to cover electrical and thermodynamic energy fundamentals in depth here; however, some related theory from the energy auditing and energy management perspective is discussed. As mentioned, the key energy consumption in manufacturing relates to electrical and thermal energy. Electrical energy powers processing equipment, pumps and motors (single and three-phase); lighting; IT equipment; electric ovens; refrigeration and HVAC equipment.

The efficiency of electricity production has increased with the introduction of efficient gas fired turbine technologies. For example, in Ireland, the efficiency of electricity generation has increased from a typical efficiency of less than 35% prior to 2000 to an average efficiency of 50%–55% currently (SEAI, 2020). Electricity transmission losses are proportional to the current squared (I^2R dissipation losses), and to mitigate these losses high voltage transmission networks are utilized. In 2015 transmission and distribution losses across European Union states ranged from 2.2% to 10.4%. Reasons for these variations include different voltage levels between member states, and differences across distribution networks (Council of European Energy Regulators, 2015). With regard to the electrical energy that is delivered to the manufacturing facility boundary, knowing how and where electrical energy is used is crucial to effective energy management. In addition, it is important to know and check the efficiency of electrical equipment regularly. For example, the efficiency of equipment such as motors can significantly drop according to changes in loading and operating characteristics.

Motors are ubiquitous in manufacturing, and range from small stepper motors to large three-phase induction and synchronous motors. Governed by the laws of electromagnetism, the rating of a motor depends on the ability to dissipate heat from I^2R losses. Small



Fig. 2 The energy hierarchy. Reproduced from Institute of Mechanical Engineers, 2020. The Energy Hierarchy: Supporting Policy Making for 'Net Zero'. [Online] Available at: <https://www.imeche.org/news/news-article/the-energy-hierarchy-a-powerful-tool-for-sustainability> (Accessed 20.01.2021).

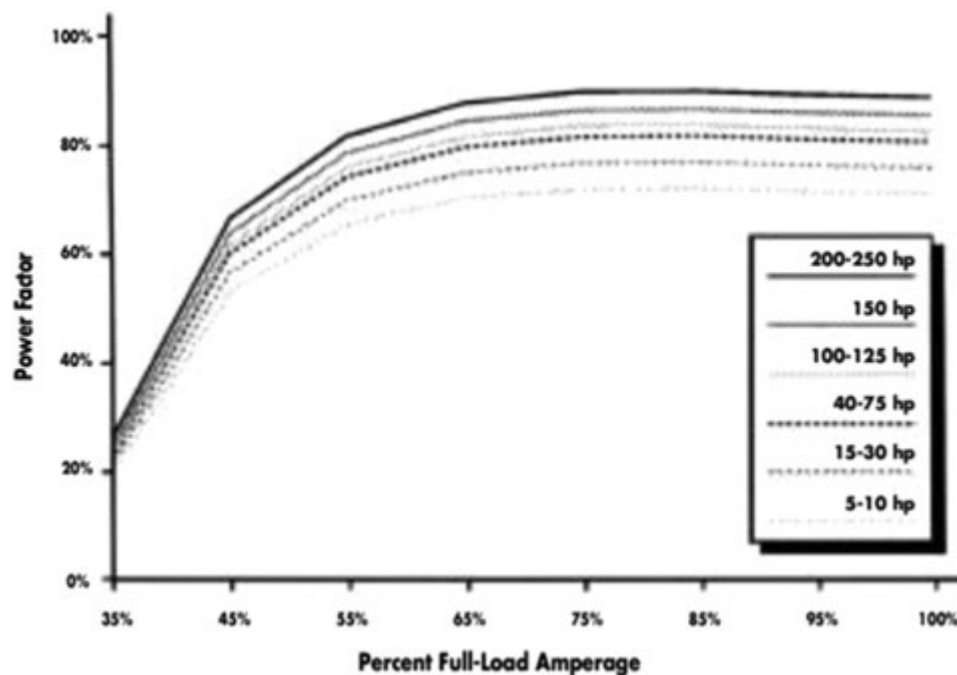


Fig. 3 Power factor versus percentage full-load. Reproduced from U.S. Department of Energy, 2014. Determining electric motor load and efficiency. [Online] Available at: <https://www.energy.gov/sites/prod/files/2014/04/f15/10097517.pdf> (Accessed 20.01.2021).

motors are inherently inefficient, whereas large motors have inherently higher efficiencies due to a relatively small resistance voltage drop term (Hughes *et al.*, 2016). Efficiency increases with speed: for a given torque, the power output is proportional to the speed, while electrical losses are generally constant. Sizing motors correctly for each application is very important to ensure that they run at a suitable percentage of their full load. Efficiency is relatively constant to approximately 75% of full-load. Efficiency then drops by approximately 5% down to 50% of full-load; however, it then drops significantly when motors operate at less than 50% of their full-load (Beggs, 2009). One other consideration for energy management is that the wide use of induction motors in industry increases reactive power, and thus can lead to poor power factors, i.e. the ratio of apparent to real/active power. Moreover, with regard to induction motors, when operating at low full-load percentages, the power factor can drop significantly. Fig. 3 shows that this pertains to a wide range of motor sizes; for various motor sizes, the power factors have dropped to below 0.3 at 35% of full-load. Poor power factor in turn can lead to higher energy charges from the utility provider, and therefore must be assessed and mitigated where possible. Power factor correction is achieved by using capacitors to counteract the effect of reactive loads (Beggs, 2009).

The use of variable speed drives and variable frequency drives is now commonplace to improve industrial energy efficiency. The basic principle and objective of using a variable frequency drive is to control the motor speed by controlling the voltage and frequency output. Traditionally throttling valves and other fluid control devices were used to regulate fluid flow for oversized pumps and fans and to deal with variable flows, resulting in fluid pressure drops, and consequent energy losses. Pumps and fans

used in fluid flow applications obey the fan laws, which relate the volumetric flow rate to the rotational speed, and the power of fans and pumps. The volumetric flow rate for a fixed blade diameter pump/fan, is proportional to the rotational speed. The power, however, is proportional to the rotational speed cubed. Controlling the rotational speed of the motor using variable frequency drives, as opposed to throttling flows, means that energy losses can be avoided and the power requirements considerably reduced for application with variable flowrates. Variable frequency drives are not required for constant flow rates providing devices are correctly sized. Furthermore, the ratio between static and dynamic head should always be assessed for a given flow system: variable frequency drives are better suited to systems with predominant dynamic head losses.

Correct sizing of equipment and plant is imperative for effective energy management: margins of safety and variation in demand can lead to uncertainty and incorrect sizing of equipment. Plant and equipment that are oversized result in increased capital costs, and lifetime efficiency losses caused by operating below optimal capacity (under-utilization). The same holds true for under-sized systems but for different reasons. In that case, the focus on minimizing capital costs, common at the start of many construction projects, can result in under-sized plant, e.g., piping and ducting, which leads to higher major head losses, resulting in greater and pumping and fan energy requirements, and higher energy costs over the lifetime of the facility.

Thermodynamic analyses are critical to understanding and optimizing industrial process energy flows. Mass conservation and the first law of thermodynamics enable the determination of first law thermodynamic efficiency. For example, the coefficient of performance of heat pumps and refrigerators is calculated as the ratio of the useful heat (evaporator heat transfer for refrigerators and condenser heat transfer for heat pumps) to the power input. Second law analyses take account of the fact that energy has both a quality aspect and a quantity aspect.

Exergy is a thermodynamic property that is derived by combining the first and second laws of thermodynamics; it quantifies the potential of a system to do useful work relative to its environment, or defined dead state. In exergy analysis convention, the dead state is often defined as standard temperature and pressure; however, the use of standard temperature and pressure may not always be useful, and the implications of doing so should be assessed on a case by case basis. Once a system reaches equilibrium with its dead state, the opportunity to do work no longer exists and the exergy is zero. The property exergy is used in an exergy balance to quantify inherent process irreversibilities. Undertaking an exergy analysis typically involves several steps for open systems: calculating the exergy of various process or systems inputs and outputs; calculating the rate of exergy destruction using an exergy balance; using the exergy balance and exergy destruction rates to calculate the exergetic efficiency of: (1) individual components within processes, (2) processes, and (3) overall process or system efficiency. Although the rate of exergy destruction, initially, may not be significant as a stand-alone quantity, it does provide a critical benchmarking tool, both for the components within a multi-component process and between similar processes. The rate of exergy destruction is also an ideal platform for assessing possible process plant improvements and optimization. There are several definitions of exergetic efficiency: the simple exergetic efficiency relates the exergy output to the exergy input; other definitions consider the ratio of the useful exergy output to the exergy input, termed the rational thermodynamic efficiency.

Exergy analyses, as the name suggests, use exergy as the basis for assessing and optimizing system efficiency. The application of exergy analysis is far reaching: cryogenics, power cycles, chemical processes, industrial and agricultural applications, and desalination (Sciubba and Wall, 2007). It is widely used and accepted by leading energy experts as providing a powerful basis for characterizing and optimizing energy and thermal systems (Fitzsimons, 2011). Exergy can be categorized as thermo-mechanical (or physical exergy) and chemical exergy. Thermo-mechanical exergy relates to the ability to do useful work as a result of system work and heat interactions with the dead state. Chemical exergy is decidedly more complicated, and relates to both the intrinsic exergy bound in substances, determined using the Szargut reference environment, and differences in exergy that arise from differences in chemical potential, according to the Gibbs energy equation. A thermo-mechanical dead state (or restricted dead state) is used as an intermediary step for the determination of differences in chemical potential. That is, the differences in thermo-mechanical energy are first determined, and then the chemical exergy is calculated, as a function of the difference in chemical potential evaluated at dead state temperature and pressure. An in-depth discussion of chemical exergy is again beyond the scope of this article. However, the specific thermo-mechanical exergy equation, and open system, steady state exergy balance equations are presented. The specific exergy, e , is defined as follows:

$$e = h - h_0 - T_0(s - s_0) + \frac{1}{2}(V^2 - V_0^2) + g(z - z_0) \quad (1)$$

In eq. 1, h is the specific enthalpy, h_0 is the specific enthalpy at the defined dead state, T_0 is the dead state temperature, s is the specific entropy, and s_0 is the specific entropy at the defined dead state. The remaining terms refer to kinetic and potential exergy where V is the velocity at thermodynamic state, V_0 is the velocity at the dead state, g is the gravitational acceleration, z is the elevation at the thermodynamic state and z_0 is the elevation at the defined dead state. The steady state exergy balance equation for open systems is shown in eq. 2:

$$\sum_i \dot{Q}_i \left(1 - \frac{T_0}{T_i}\right) + \sum_i \dot{W}_i + \sum_{in} \dot{m}_i e_i - \sum_{out} \dot{m}_i e_i - \dot{E}_D = 0 \quad (2)$$

where $\dot{Q}_i$ relates to the rates of exergy transfer associated with any heat transfers which may take place at various locations i on the system boundary where the instantaneous temperature is T_i . The second term relates to the rates of exergy transfer associated with work transfer, $\dot{W}_i$. The third and fourth terms relate to the relevant transfers of specific exergy e_i into and out of the system by mass flow $\dot{m}_i$, and $\dot{E}_D$ refers to the rate of exergy destruction.

Exergy has been used as a means to investigate the energy performance of manufacturing processes, where the authors developed a framework to thermodynamically characterize the material and energy resources used in multiple manufacturing processes (Gutowski *et al.*, 2008). Energy performance data for 20 manufacturing processes were collated from the literature and analysed; process energy intensity data (joules per kilogramme of material processed) versus processing rate data (kilogramme of material processed per hour) were plotted for each of the 20 processes. These processes ranged from traditional processes such as machining, casting, and injection molding, to more technically-advanced processes such as electrical discharge machining, and the vapor phase processes used in the semiconductor industry and nanomaterials processing. One of the key findings was that the energy intensity of materials processing had increased significantly over the past number of decades, by at least several orders of magnitude. The processes at the higher energy intensity levels included thermal oxidation processes for semiconductor processes and electrical discharge machining drilling. Medium intensity processes again included several processes typical of complex manufacturing processes, for example, chemical vapor deposition and dry etching. Low energy intensity processes included well-established and relatively high throughput processes such as injection molding, machining and melting. The authors concluded that many of the newer, high-tech manufacturing processes used “high exergy value materials in very inefficient ways (Gutowski *et al.*, 2008)”. Although acknowledging the benefits of these manufactured products, including environmental benefits by means of longer life or lower use-phase energy requirements, the authors nevertheless sounded a timely alarm regarding the often wasteful use of energy and materials in newer, advanced manufacturing processes.

More recently, and following a similar approach to their earlier work (Gutowski *et al.*, 2008), the energy intensity of several additive manufacturing processes was collated and plotted alongside the original 20 processes, see Fig. 4 (Gutowski *et al.*, 2017). With regard to the additive manufacturing processes, it was found that:

- (1) There was a wide range of process types and energy intensity values associated with additive manufacturing processes
- (2) In general, additive manufacturing processes have lower power requirements compared to the majority of conventional manufacturing processes
- (3) In general, the additive manufacturing processes had lower processing rates than conventional processes (kilogramme of material processed per hour)
- (4) The additive manufacturing processes had higher specific energy values or intensities (joules per kilogramme of material processed) than most of the traditional processes. (Gutowski *et al.*, 2017)

For a more complete life cycle energy analysis, consideration should be given also to the energy related to the part post processing as well as the produced product usage and end of life, relative to alternative conventional designs and production routes. Other important energy considerations are the pre-process embodied energy of the raw materials of the part composite including the matrix and reinforcement, coupled with consideration of the resulting part design resulting weight specific functional properties.

Energy Management Systems

The International Organization for Standardization (ISO) have developed two standards that relate to energy management. The original standard that addressed energy usage was the Environmental Management Standard, ISO 14001. The energy focus in ISO 14001 is primarily related to the link between energy consumption and the associated emissions (both direct and indirect emissions). However, within ISO 14001, energy is only one consideration among a number of other environmental factors including waste reduction, the sourcing and use of sustainable materials, water usage and the management of chemicals.

The Energy Management Standard, ISO 50001, deals solely with energy management. The current version of the ISO 50001 Energy Management Standard was published in 2018, superseding the earlier 2011 version. ISO 50001: 2018 is similar in structure to the other ISO family of standards, which are based on the Plan-Do-Study/Check-Act Deming cycle (The Deming Institute, 2021). Accordingly, it is a systems-based approach where the user develops and implements systems and processes to continually improve energy performance. In the standard, it is important to note that energy is broadly defined as: electricity, fuels, steam, heat, compressed air and other similar media. In the ISO 50001 terminology, there are three aspects of energy performance: energy efficiency, energy use, and energy consumption. Energy efficiency is a quantitative measure of how much energy is consumed per some operational or capital metric, for example, energy consumption per meter squared of factory floor-space or energy consumption per unit of production. Energy use relates to how energy is used, for example, heating and cooling, processing equipment, and transportation. Energy consumption, finally, pertains to the quantity of energy that is “applied” in the various energy use categories.

The standard is broken down into a number of sections, and is similar in structure to other ISO standards, setting out and acknowledging the importance of leadership, planning, and operational competencies and processes. A baseline of energy performance is determined using a suitable energy auditing methodology. Energy performance is calculated in terms of metrics, called energy performance indicators (EnPIs); these are user-defined and should be relevant to the specific context of the organization. Once the baseline is determined, the objective is to continuously improve energy performance.

Energy Auditing

An energy audit can be defined as a “process of determining the types and costs of energy use in the building, evaluating where a building or plant uses energy, and identifying opportunities to reduce consumption (Thumann *et al.*, 2013)”. The ISO auditing

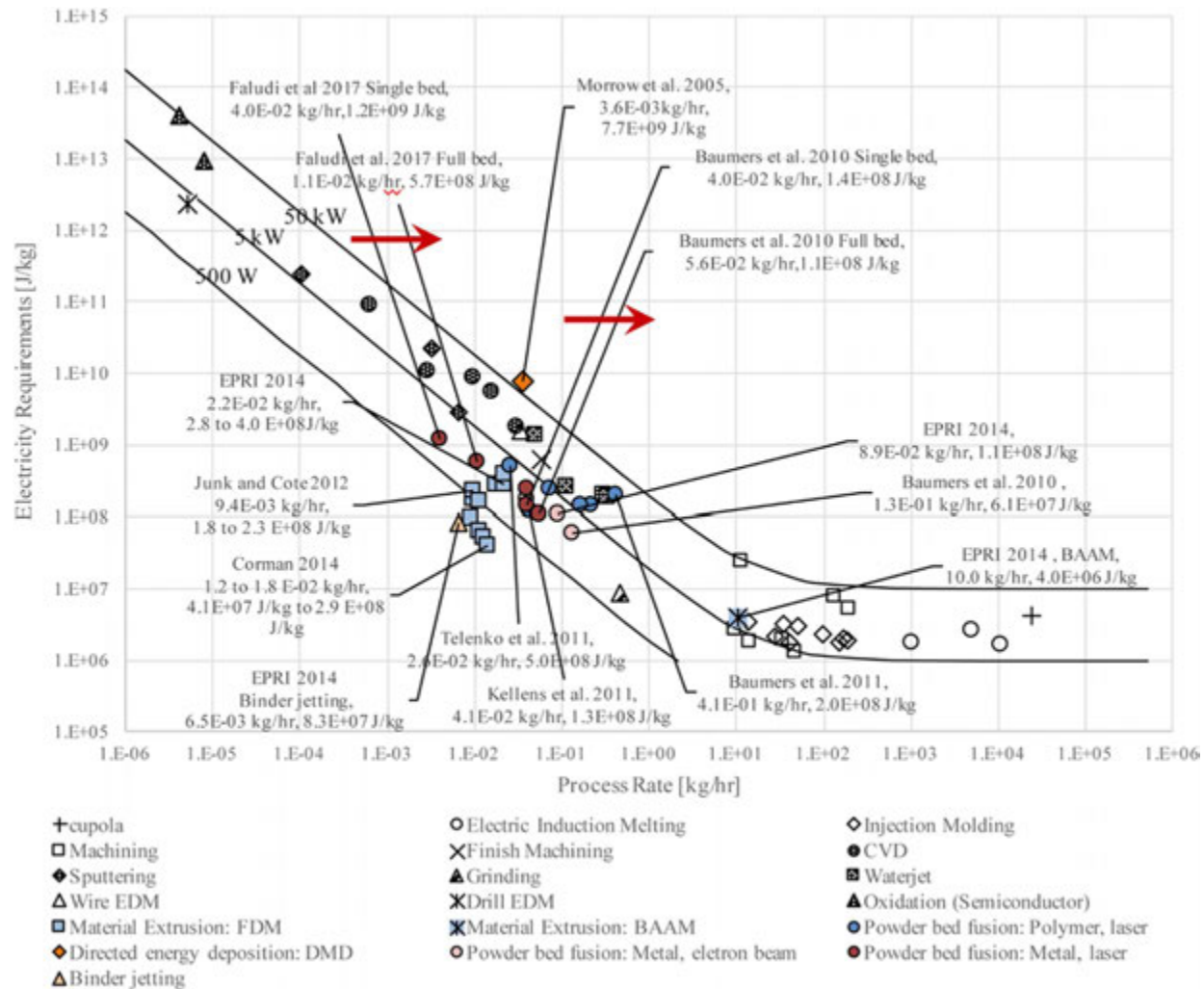


Fig. 4 Energy intensity versus processing rate for manufacturing processes. (a) Used with permission from Gutowski, T., *et al.*, 2017. Note on the rate and energy efficiency limits for additive manufacturing. *Journal of Industrial Ecology* 21 (S1), S69–S79. (b) Note that source references and supporting data are contained in both Gutowski, T., *et al.*, 2008. Thermodynamic analysis of resources used in manufacturing processes. *Environmental Science and Technology* 43 (5), 1584–1590.

standard, ISO 50002 Energy Audits, aligns with ISO 50001 and describes audit outputs as providing information on current use and performance, and as a means to rank recommendations for improvement in terms of energy performance and economic benefits (International Organization for Standardization, 2014). The Energy Audits standard provides a framework for the auditing process, defines the competency requirements of the energy auditor, and provides guidelines for conducting, analysing and reporting energy audits.

There are several different scopes of energy audits, from the basic assessment of electricity and fuel bills to a comprehensive auditing plan to obtain accurate measurements, followed by detailed analyses and modeling of facilities, processes and devices. The old adage “you can’t manage what you can’t measure” holds true; however, there is an inherent effort and cost associated with obtaining detailed oversight of energy flows, particularly in terms of personnel, equipment, time, operational disruption, and even process risk. As a result, it is important that each manufacturing facility determines the scope of energy audit that is relevant to them.

The American Society of Heating and Air-Conditioning Engineers (ASHRAE, 2021) and the *Handbook of Energy Audits* report different energy auditing scopes, although they are analogous. The ASHRAE energy auditing proposes a preliminary assessment, followed by three levels of energy audit:

- (1) Preliminary assessment: historic energy cost data and determining an energy utilization index.
- (2) Level 1 - walk-through and brief survey.
- (3) Level 2 - energy survey and analysis (breakdown of energy usage and cost; potential improvement projects including projects requiring capital investment; accounts for the people factor; maintenance procedures).

- (4) Level 3 - detailed analysis, including economic analysis of projects (detailed data gathering and analysis; project cost and savings analysis).

In a similar manner, the *Handbook of Energy Audits* suggests and discusses four types of energy audit (Thumann *et al.*, 2013):

- (1) Type 0 – the Benchmark Audit, which is analogous to the ASHRAE preliminary assessment in that it includes an analysis of energy costs and determining key performance energy indicators and metrics.
- (2) Type I – the Walk-through Audit concerns a tour of the facility and initial comparison with industry averages.
- (3) Type II – the Standard Audit quantifies energy usage and losses; focuses on equipment and operational characteristics; efficiencies are determined and potential cost savings based on proposed improvements are quantified.
- (4) Type III – Computer Simulation provides detailed breakdown of energy use baseline and usage patterns; simulation to predict energy consumption; considers system interactions.

From the energy auditor's perspective, internal or external, the initial objectives in the preliminary stages of the energy audit process are to gain an understanding of the organization's energy use. Some important things to consider and review include:

- (1) The business function and primary objectives
- (2) The regulatory landscape (e.g., Food Safety, Pharmaceutical, Environmental)
- (3) The production levels (variable or constant)
- (4) The production lines (multiple, variable products or consistent product line)
- (5) The manufacturing processes (complex or relatively simple)
- (6) The specific processes, equipment and instrumentation that is used
- (7) Utility bills (current and historic – of sufficient duration to cover seasonal and other fluctuations)
- (8) Electricity tariffs (i.e., night rate options, power factor charges).

The second step is the site visit, which includes a walk-through of plant and processes. This provides an opportunity to talk with the relevant engineers, operators and managers about specific facilities and processes: HVAC, steam heating, specialized manufacturing equipment, cleanrooms, water treatment. Other important information is reviewed and collated, and requested if not available:

- (1) Process flow diagrams, P&IDs, equipment schematics if available.
- (2) Control setpoints.
- (3) Instrumentation calibration protocols, maintenance schedules for equipment etc.
- (4) Process inputs, process outputs, process loops, waste streams.
- (5) Process layout.
- (6) Location of electrical/control panels, instrumentation etc.
- (7) Electrical and control panel accessibility.

The focus of the energy audit will typically include the building envelope, the electrical system and the HVAC system, as well as the specific processing equipment. Determining the breakdown of energy consumption at sufficient resolution is critical to ensure that energy efficiency improvement areas are correctly targeted. To this effectively, a detailed measurement plan should be drawn up, determined by what is currently being measured, and the frequency and accuracy of the current measurements. Activities may include identifying and/or assessing the following:

- (1) Systems/sub-systems that may require detailed monitoring.
- (2) The type of instrumentation that is available and/or required: e.g., energy meters, pressure, flow, temperature, level etc.
- (3) The parameters that should be measured.
- (4) The accuracy and calibration status of instrumentation.
- (5) The applicability of the instrumentation (e.g., some energy meters measure currents greater than 3 A).
- (6) The required sampling frequency and duration.

Energy auditing instrumentation requirements are largely driven by the type and location of the manufacturing plant. Electrical power measurement is common to all manufacturing plants. There is a wide range of power and electrical measurement devices available, from simple and cost effective clamp on, or plug in, meters to sophisticated power quality analyzers. Power quality analyzers provide detailed information on multiple electrical power parameters including power factor, harmonics, and some devices incorporate energy loss calculation algorithms (FLUKE website, 2021). Other commonly used measurement equipment includes gas analyzers for assessing the efficiency of combustion equipment; flow measurement devices for liquids and gases; and pressure, temperature, humidity and air velocity measurement devices. Please see Thumann *et al.* (2013) for detailed coverage of energy audit instrumentation. Determining the sampling frequency depends on a number of factors: the variability of the data and, realistically, the data storage capacity of the measurement device. Ideally, highly varying data requires high frequency data capture. However, at times a compromise may be necessary between acceptable sampling frequency and data storage limitations. With regard to the sampling duration of the energy audit, it should be representative and again capture any data variability, for example: seasonality, production levels, production variability, production shifts etc.

The retrieved measurement data will require at least basic data processing, such as data smoothing and the removal of outliers. Once done, the data is collated, analysed, and reported as relevant energy metrics. The selection of suitable energy metrics or key

performance indicators results from the earlier consideration of the business function and objectives. Based on the assessment of energy performance, it is usually possible to make recommendations for energy use and efficiency improvements, and to quantify preliminary potential savings. Sometimes this may be very obvious, such as processing equipment that is left on or fluid leaks e.g., compressed air. However, other times it can be less so; an energy metric in isolation can appear meaningless. To provide context, benchmarking, is a very useful approach that facilitates the tracking and assessment of energy performance, both internally (relative to past performance) and externally (relative to peers).

Energy Star is U.S. based, industry specific, benchmarking initiative that enables energy performance assessment and comparison within relevant industrial sectors. According to the Energy Star website, benchmarking is a critical step in energy management (Energy Star, 2021). The Energy Star scoring system enables manufacturing plants to compute a single “energy score”, on a percentile basis, in order to assess energy performance and obtain certification. Using survey data collected from various industrial sectors, energy performance regression models were developed for each of those specific sectors (e.g., automobile, glass, steel, cement, aluminum casting). User data is entered in the Energy Star spreadsheet, which takes into account, and adjusts for, specific key variables relevant to the industrial sector. Using the entered data, an energy score is computed. This percentile-based score enables the organization to assess their energy performance relevant to their industry peers. Manufacturing plants that earn an energy score of 75 or higher eligible to earn the ENERGY STAR certification for superior energy performance (Energy Star, 2021).

Energy management often crosses several functional boundaries within an organization, and therefore it is important to assess energy performance at the process, system and plant levels. Input from personnel across the relevant organizational functions in the energy management and auditing team contributes towards a broader outlook. Furthermore, understanding both the demand side and supply side of energy management provides opportunities for synergy. Ideally, the supply of utilities (e.g. process water, compressed air, heating and cooling) should be as energy efficient as possible. However, the end-users of the utilities should be aware of the energy intensity of the utilities they use. In manufacturing settings, there is a general need to challenge and understand the rationale for the quality levels of given utilities, and to conserve those utilities where possible. Quality decisions are often outside the control of the manufacturing facility, driven by regulatory bodies who impose highly risk-averse quality requirements, ultimately resulting in increased energy consumption. Moreover, quality ‘rules of thumb’ can exist in various sectors, for example, the frequency of air change rates in cleanrooms, or process water quality requirements in the semiconductor industry and pharmaceutical industries. In critical manufacturing industries, quality and safety is paramount; however, quality requirements that impact significantly on energy consumption should be assessed and justified with scientific evidence.

Notwithstanding the effectiveness of energy auditing as a key tool for energy management, embedding energy efficiency at the design phase should be the preferred option. Designing for energy efficiency includes matching capacity to demand, choosing the best available technology in terms of energy efficiency for a given process, and also ensuring that on-line or off-line energy measurement equipment and instrumentation is integrated into the manufacturing facilities and SCADA systems. Planning for energy efficiency at the outset facilitates better energy management in the longer term.

Conclusions

This article has presented a review of energy related to industrial processing methods and the assessment methods with consideration that they could be applied for the analysis of composite materials production. Effective energy management and efficient energy use are crucial to sustainability efforts. Recent IEA energy data do not inspire confidence with regard to the energy-related efforts to mitigate climate change. Energy considerations are pervasive throughout the composites life cycle, from the extraction of raw materials to the processing and facilities energy requirements in composites manufacturing. The energy intensity of manufacturing processes has reportedly increased by several orders of magnitude over recent decades, with advanced manufacturing processes typical of the semiconductor and nanomaterials processing sectors being particularly energy intensive. Energy management standards and energy auditing play important roles in overall energy management initiatives. Relevant energy methodologies and holistic environmental impact tools, such as exergy analysis and Life Cycle Analysis, identify key impacts and efficiency opportunities. However, given the urgent need to reduce energy in absolute terms, and the current energy trajectory, it is unlikely these efforts will suffice.

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Additive Manufacturing for Sustainability of Composite Materials Production

Eanna McCarthy and Dermot Brabazon, I-Form, Advanced Manufacturing Research Centre, and Advanced Processing Technology Research Centre, School of Mechanical and Manufacturing Engineering, Dublin City University, Dublin, Ireland

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Introduction

Additive manufacturing (AM) is a class of advanced manufacturing process where a three-dimensional computer model is reproduced as a physical part by adding material layer by layer (ASTM International, 2012). AM initially saw use as a method of rapidly producing models and prototypes based on digital designs, with its low lead time allowing for parts to move quickly from design to physical model, and be easily updated with revisions or customizations. Increasingly, AM is used as a manufacturing method in its own right. The annual market worth growth rate for AM has been reported as 26.2%, reaching \$5.165 billion in 2015, with revenues projected to exceed \$21 billion worldwide by 2020 (uz Zaman *et al.*, 2018; Attaran, 2017).

While AM methods are typically less cost-effective for mass production than conventional manufacturing methods, there are a number of key advantages that have allowed AM to become established as a technique for use in direct or indirect manufacturing of commercial parts. AM allows for complex topology-optimized structures which can deliver high strength-to-weight ratios, provides the ability to revise or customize parts without adjustments to the production setup, allows for parts to be printed to order, and can reduce waste compared to subtractive manufacturing (Dietrich *et al.*, 2016). Many of these advantages can provide improved sustainability, such as increased fuel efficiencies from light-weight parts, reducing or eliminating storage costs and obsolescence, and reduced material waste. Verhoef *et al.* estimate global energy savings due to AM of 5%–27% are achievable by 2050, primarily from reduced material usage and transport energy usage (Verhoef *et al.*, 2018).

Composite materials are materials made up of two distinct constituent materials with differing properties and can provide more sustainable material solutions. For example, their ability to deliver high strength-to-weight ratios that produce lightweight parts can improve fuel efficiencies in aerospace and automotive applications (Cheon *et al.*, 1997; Macke *et al.*, 2012; Joo *et al.*, 2020; Poulidikidou *et al.*, 2016). Using additive manufacturing to produce composite parts has the potential to combine the physical properties of each material and improve the respective sustainability of both. In this article, general sustainability of AM, general sustainability of composite materials, and the sustainability of AM of composite materials will be discussed.

Sustainability in Additive Manufacturing

Sustainability is the potential for something to continue indefinitely. For industry improving sustainability means reducing material and energy consumption, using renewable sources of material and energy, making use of recycling, and minimising or eliminating impact to the environment from emissions, hazardous chemicals etc. Examples of improvements to sustainability are creating longer life-time parts, reducing obsolescence, creating light-weight parts which can improve fuel efficiencies and reduce transport impacts, green energy production which makes use of sustainable sources and removes dependence on fossil fuels, using recycled materials to make parts, and making parts which can be recycled.

Additive manufacturing methods have a number of sustainability benefits. Additive manufacturing methods are capable of creating complex geometries that are difficult, or impossible, to produce with conventional methods. In particular, there has been emphasis on topology optimized or latticed parts, where design requirements are achieved with the minimum amount of material, which are possible with AM's design freedom (Emmelmann *et al.*, 2011; Yang *et al.*, 2016; Xu *et al.*, 2019; Chantarapanich *et al.*, 2014).

With subtractive manufacturing or forming methods, there is limited access to the interior structure of parts, as tool-paths are blocked by the exterior structures of the part. In AM as the part is built up layer by layer this issue is eliminated. By minimising the material required in a given part, topology optimized lattice parts improve sustainability in two ways: reducing overall material usage in manufacturing, and creating lighter parts which can improve fuel efficiencies in automotive and aerospace applications.

For example, Primo *et al.* present the integration of topology optimisation into additive manufacturing of a simple C-clip (Primo *et al.*, 2017). The topology optimized and latticed clips produced using a Solidscape 3Z STUDIO material-jetting-based AM machine are presented in Fig. 1. In this work, possible designs were evaluated against the Key Performance Indicators (KPIs) displacement, weight, stress, and printing time, the acceptable ranges for which these may vary based on the manufacturer or customer requirements.

Similar approaches have been taken with aerospace parts. Seabra *et al.* applied topology optimisation to an aircraft bracket (Seabra *et al.*, 2016). They report a reduction in part volume of 54% in the part volume from the original bracket design (see Fig. 2). The material was changed from aluminum to titanium, resulting in a weight decrease of 28% with twice the original safety factor.

Topology optimized aircraft parts have also been investigated commercially. The European Aeronautic Defense and Space Company (EADS) Innovation Works developed topology optimized aircraft parts manufactured with Direct Metal Laser



Fig. 1 Additively manufactured C-clips with different topology optimized lattice structures to minimize material usage while meeting KPIs. Reproduced from Primo, T., Calabrese, M., Del Prete, A., Anglani, A., 2017. Additive manufacturing integration with topology optimization methodology for innovative product design. *Int. J. Adv. Manuf. Technol.* 93 (1–4), 467–479.

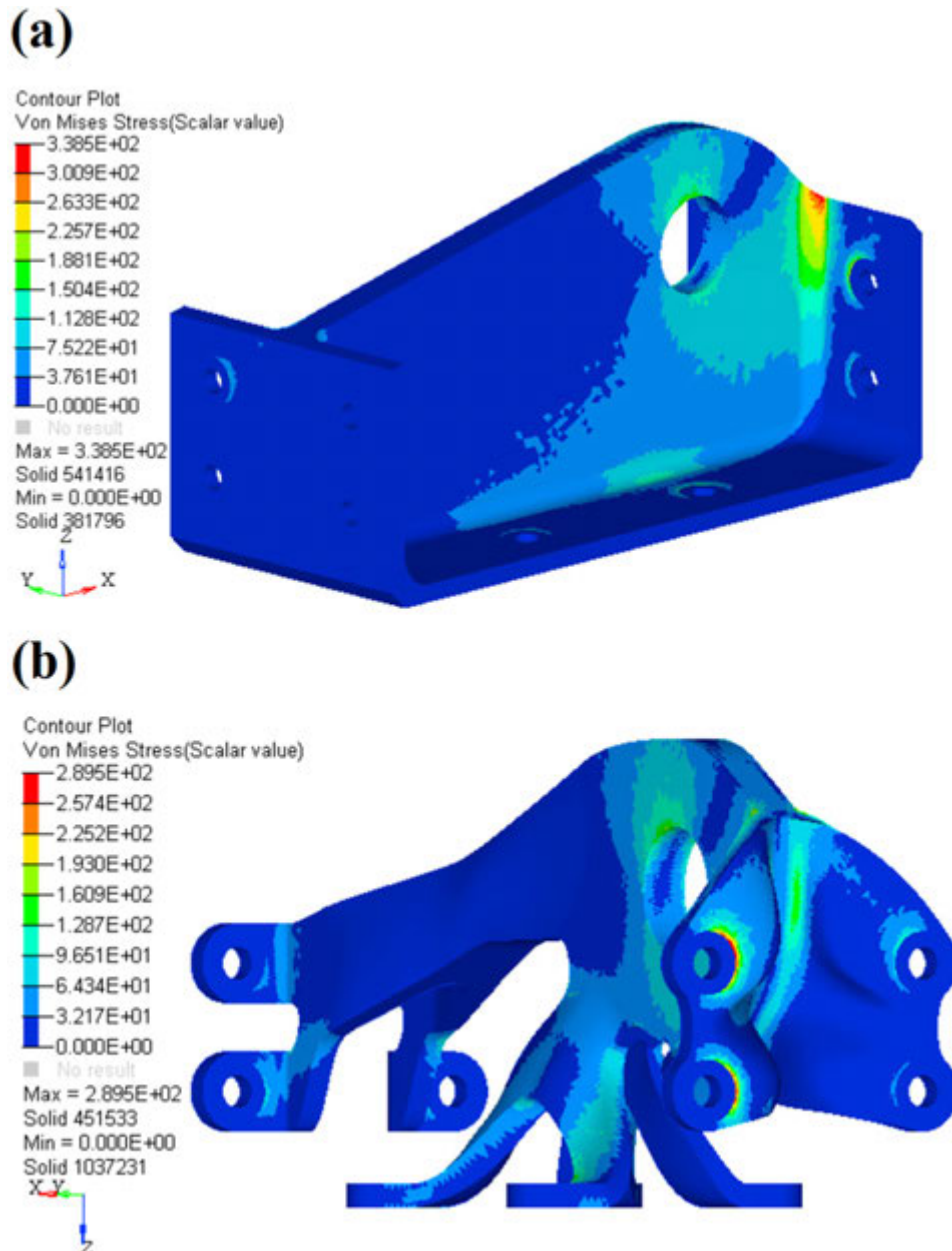


Fig. 2 Stress distribution in (a) original design for an aircraft bracket, and (b) topologically optimized design for the bracket. Reproduced from Seabra, M., Azevedo, J., Araújo, A., *et al.*, 2016. Selective laser melting (SLM) and topology optimization for lighter aerospace components. *Procedia Struct. Integr.* 1, 289–296.

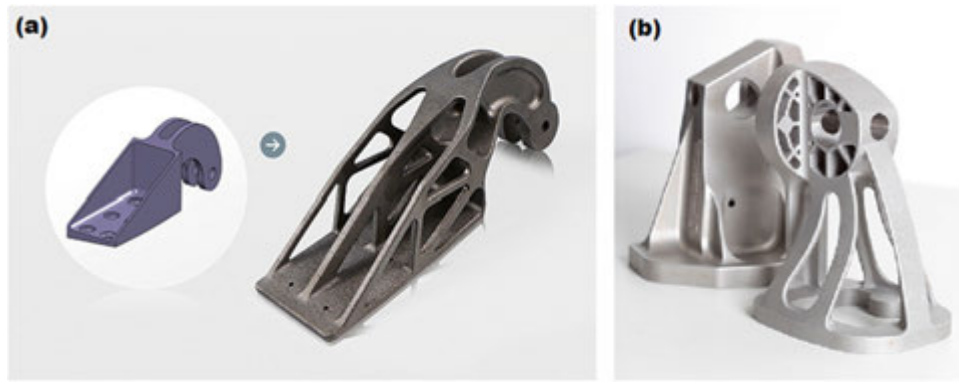


Fig. 3 EADS Innovation Works developed topology optimized design aircraft hinge brackets manufactured using DMLS: (a) Conventional bracket design (left) and topology optimized titanium DMLS bracket (right), (b) Conventional bracket (rear) and topology optimized stainless steel DMLS bracket (front). Reproduced from EOS GmbH, 2020. Aerospace: EADS and EOS - Study demonstrates savings potential for DMLS in the aerospace industry. [Online]. Available: https://www.eos.info/press/customer_case_studies/eads. (accessed 30.03.2020).

Sintering (DMLS) (EOS GmbH, 2020). **Fig. 3** shows conventional and topology optimized AM-produced Airbus hinge brackets. Material usage reduced via topology optimisation leads to a 40% reduction in energy usage over the lifecycle of the part (manufacturing and operation), despite higher energy usage for the AM process. Their study suggested that this topology optimisation could reduce the weight of the plane by 10 kg. The CO₂ emissions for the hinge could be reduced by 40% over the total life-cycle of the part, and the raw materials used were reduced by 25% compared with using rapid investment casting. By reducing energy/fuel consumption and corresponding CO₂ emissions, and reducing material consumption, these optimized lightweight part designs can make a significant contribution to sustainability. Huang *et al.* estimate potential energy savings for the aerospace industry of 70–173 million GJ/year, and a cumulative CO₂ emission reduction of 93–215 million tons per year by 2050, by use of lightweight AM parts (Huang *et al.*, 2016).

The reduced lead times and lack of specific tooling for additive manufacturing can allow parts to be quickly and easily printed to order. This reduces the need for storage costs, and associated energy use, by allowing spare parts to be produced quickly using AM and put directly into use. Transport fuel/energy use may also be reduced by having parts produced locally with interchangeable AM machines, rather than produced centrally with a specific production line and shipped. As such, files are sent electronically rather than parts sent physically. The AM production can be carried out by a local branch of the manufacturer, a local third-party AM center, or even by the customers themselves.

Ryan and Eysers categorized the to-order production of automotive spare parts into five scenarios: Personal Manufacturing - Customers printing parts with consumer desktop AM machines; Retail Manufacturing - Parts being additively manufactured by garages, workshops, or dealerships; Bureau Manufacturing - Third-party AM centers being licensed to produce parts; Factory Manufacturing - AM being used to produce spare parts at the factory; and Mobile Manufacturing - parts being produced by AM in transit, turning the lost time of transport into productive time (Ryan and Eysers, 2017). The potential benefits the authors propose for each scenario is presented in **Table 1**. The possible sustainability benefits include reducing storage and transport needs, reducing packaging, and reducing obsolescence. If spare parts are out-of-production, when the existing stock of parts is depleted vehicles, products, and machinery that may be otherwise functional and fulfilling a need may become obsolete and be removed from use. With to-order production via additive manufacturing, obsolescence can be delayed. Siemens report a commercial example, in which a metal impeller for a fire protection water pump in a nuclear power plant in Krško, Slovenia was replaced with a AM-produced replica (Siemens, 2020). The original part, produced in 1981, was no longer in production and spares were unavailable. A digital “twin” of the original part was created, and an AM replacement produced, allowing the continued operation of the plant. Increasing lifetimes for products and equipment by delaying obsolescence is a significant sustainability benefit which AM can provide.

Verhoef *et al.* present a case study examining the reduction of transport costs/emissions using local AM (Verhoef *et al.*, 2018). The authors predicted the transport energy usage for manufacture of composite panels for the Airbus A320 in 2050 (see **Table 2**). For conventional production, aluminum is transported from Pittsburgh to Taiwan where the composite panels are produced and then shipped to Toulouse for assembly. The authors suggest producing the panels locally with AM, eliminating the transport energy use of shipping to Taiwan, and making an energy saving of 1.22 PJ a year.

Sustainability in Composites

Improved Properties

Composite materials offer an opportunity to improve sustainability by providing desirable properties, such as improved wear resistance or high strength-to-weight ratios, which can lengthen part lifetimes or improve fuel efficiencies in automotive/aerospace

Table 1 Analysis of potential benefits of AM for five spare part production scenarios

Potential benefits of AM	Scenario 1: Personal manufacturing	Scenario 2: Retail manufacturing	Scenario 3: Bureau manufacturing	Scenario 4: Factory manufacturing	Scenario 5: Mobile manufacturing
Distributed Manufacturing	X				X
Reduced Inventory		X			X
Centralized Manufacturing			X	X	
On-Demand Manufacturing	X	X	X	X	X
Reduced Transportation	X	X			
Better Responsiveness	X	X	X		
Low Volume Manufacturing	X	X	X		
Manufacturing in Remote Locations	X				X
Reduced Overall Costs			X	X	
Sustainability Improvements	X	X			
Improved Customer Satisfaction	X	X	X	X	X
Reduced Obsolescence Risk	X	X	X	X	X
Reduced Downtime			X	X	X
Better Flexibility		X	X	X	
Robustness to Supply Chain Disruption		X	X		
Production Capacity Buffer		X			
Reduced Packaging	X				X
Manufacturing Postponement	X	X	X	X	X
Reduced Parts Shortage	X	X	X	X	X
Produce Obsolete Parts	X	X	X	X	X

Note: Reproduced from Ryan, M.J., Evers, D.R., 2017. Digital manufacturing for spare parts: Scenarios for the automotive supply chain. In: Proceedings of the Third Summit ACMA Centre for Technology.

Table 2 Predicted transport energy usage and reduction for moving to local AM production of composite panels for Airbus A320 in 2050 case study

Airbus A320 Transport	Trajectory	Distance (km)	Weight (million kg)	Transport Energy (PJ)
Without AM	Pittsburgh - Taiwan	12,400	2,720	2.19
	Taiwan - Toulouse	10,300	272	0.18
Total				2.37
With AM	Pittsburgh - Toulouse	6,500	2,720	1.15
Energy savings				1.22

Note: Reproduced from Verhoef, L.A., Budde, B.W., Chockalingam, C., García Nodar, B., van Wijk, A.J.M., 2018. The effect of additive manufacturing on global energy demand: an assessment using a bottom-up approach. Energy Policy 112 (October), 349–360.

applications (Cheon *et al.*, 1997; Macke *et al.*, 2012). Composites are made up of two or more materials with differing advantageous properties. Fibre-Reinforced Plastics (FRPs), fibres such as glass or carbon in a polymer matrix, provide high specific stiffness, specific strength, impact strength, and damping (Cheon *et al.*, 1997). The high specific strength makes FRPs attractive materials for lightweight parts in transport applications, where decreasing weight is a major motivator to improve fuel efficiencies and meet new targets for carbon emissions (Joo *et al.*, 2020). In automobiles, FRPs are a popular choice for interior and exterior fixtures/panels. For example, heavy truck bodies which were formerly made of steel have been largely replaced by composites, decreasing truck weight and increasing fuel efficiency and thus sustainability (Poulakidou *et al.*, 2016). Cheon *et al.* report on composite side-door impact beams, which they found could resist comparable external loads to high strength steel with a 30% reduction in weight (Cheon *et al.*, 1997). In aerospace, approximately 50% of the airframe of aircrafts are made of composite materials of some kind (Yakout and Elbestawi, 2017).

Composites are also used in wind turbine blades (Brøndsted *et al.*, 2005; Su and Kam, 2020; Rahimizadeh *et al.*, 2019). Wind energy offers a renewable energy source which facilitates the move away from non-renewable, carbon-emission heavy sources, like fossil fuels, towards sustainable energy production. Wind turbine blades require high stiffness to function, low weight/density to minimize gravity forces, and good fatigue life to give long part lifetimes (Brøndsted *et al.*, 2005). FRP composites, such as fiberfill, have high specific stiffness which makes them well suited for this application.

Metal Matrix Composites (MMCs), where non-metal additives are embedded in a metal material, can provide attractive engineering properties such as high tensile strengths, toughness, ductility, and wear resistance (Menezes *et al.*, 2013). Graphite in a metal matrix such as aluminum or copper can provide good lubrication and reduced wear rates which is attractive for automotive applications such as pistons, cylinder liners, and bearings (Macke *et al.*, 2012). Wear reduction can increase the lifetime of parts, improving sustainability.

The advent of nanostructured materials has led to the creation of nano-composites, where materials are reinforced with nanomaterials such as nanoparticles, nanotubes, or nanowires. Nano-composites can have different matrix types, such as metals and polymers, and can achieve improvements in the strength, hardness, stiffness, tribology, and other properties. Ibrahim *et al.* report on nickel-aluminum based nano-composites for use in aerospace bearings (Ibrahim *et al.*, 2019). The authors powder-sintered composites of NiAl with 5 wt% vanadium oxide (V_2O_5) nanowires and 1.5 wt% graphene nano-platelets, and achieved significant improvements in the wear resistance at various sliding speeds. Bansal *et al.* report on polymer matrix nano-composite of epoxy (bisphenol-A) reinforced with graphene oxide (Bansal *et al.*, 2018). The nano-reinforcement gave improvements to the hardness and fracture behavior.

Recycled Materials and Recycling of Composites

Polymer matrix composites

Their wide-range of advantageous properties give composites benefits to sustainability, but the sustainability of the composites themselves, their production, and disposal must also be considered. While FRPs in wind turbine blades contribute to sustainable energy production and in automotive/aerospace applications contribute to fuel efficiency, the common FRPs that can be used are costly and can be difficult to recycle, driving interest in identifying biodegradable or recyclable composites (Cherrington *et al.*, 2012; Ayre, 2018). Wood-polymer composites are a possible eco-friendly alternative (Lieblein *et al.*, 1982; Huang *et al.*, 2015). Bamboo is an attractive sustainable resource for these composites, due to its low cost, fast growth rate, high stiffness and strength, and ease of workability (Huang *et al.*, 2015; Zhang *et al.*, 2013). Huang *et al.* evaluated the properties of bamboo-epoxy and other wood-epoxy laminate composites for use in wind turbine blades (Huang *et al.*, 2015). They report that the wood based composites generally have lower specific stiffness ($13.9\text{--}22.4\text{ GPa cm}^3/\text{g}$) than conventional FRP composites ($20.5\text{--}89.9\text{ GPa cm}^3/\text{g}$), with only the *betula*-epoxy laminate out-performing the glass-polyester or glass-epoxy laminates. While the performance may be lower, the authors estimate that one imperial ton of the bamboo-epoxy composite material would cost 5% of the cost for one ton of the carbon fibre-epoxy composite material, and be more environmentally-friendly/sustainable.

Hermansson *et al.* carried out life cycle assessments of lignin based carbon fibre reinforced polymer composites (CFRPs) using recycled carbon fibres (Hermansson *et al.*, 2019). Lignin is a complex aromatic biopolymer which is abundant, biodegradable, and often available in waste materials (Haq *et al.*, 2020; Zhang *et al.*, 2019). The authors note that a shift to CFRP over other materials does not necessarily generate environmental improvements, with an increase in life cycle energy in more than half of cases examined, due to the high energy requirements for producing carbon fibre. To offset the environmental costs of producing the carbon fibre, the lifetime of the composites or the fibres themselves must be maximized. Recycling of fibres is one way to achieve this. The use of ecologically friendly materials like lignin as a matrix can also offset the environmental costs of the fibres for CFRPs.

Recycling of composites is dependent on the constituent materials. Thermoplastics, for example, are a possible matrix material and are well-suited to recycling as they can be heated and reshaped, while thermosets which must be chemically broken down and repolymerized are very difficult and costly to recycle. However even if one or more of the constituents is easily recyclable, the mixing of the materials in the composite may necessitate extra steps to separate the materials, complicating the recycling process. For carbon fibre polymer composites, pyrolysis or solvolysis may be used to reclaim the expensive carbon fibres for reuse, with the polymer matrix being broken down by heat or solvents (Meng *et al.*, 2017; Oliveux *et al.*, 2017).

Metal matrix composites

As with polymer composites, there is interest in recycling of MMCs to improve sustainability and reduce waste, and to amortize the costs of the more expensive materials and processes over longer lifetimes (Yang *et al.*, 2012). Creating MMCs using recycled industrial or agricultural waste materials is also an attractive avenue for improving sustainability. Recycled materials have the potential to be used as matrices, reinforcements, or both.

Aluminum alloys can be reinforced with ceramic particles to create a MMC with improved tensile and yield strengths, and good strength-to-weight ratios (Geiger and Walker, 1991). Schuster *et al.* investigated the recycling of 6061 Al with Al_2O_3 or SiC reinforcement (Schuster *et al.*, 1993). Firstly, the authors looked at recycling of foundry scraps. Foundry returns from gates and risers could be fluxed and degassed using salt addition and gas injection, to remove inclusions, oxides, and other impurities, and reused in future shape-casting. The authors also report on reclamation of the matrix material from Al MMCs using rotary salt furnace technology, with 64% of the metal being recoverable.

Enginsoy *et al.* report on the use of recycled aluminum in fabrication of MMCs by sintering and forging (Enginsoy *et al.*, 2020). The aluminum alloy used, AA7075, is widely used in aerospace and automotive applications, giving good availability of scrap for recycling. The authors used fine atomized powder produced by atomization from recycled AA7075 and pure copper, with Nb_2Al and SiC particle reinforcement. The combined sintering-forging method provided good results, and is economic and industrially scalable.

Fly ash is a powder composed of spherical ash particles, composed mainly of silicate, aluminate, and calcium compounds, produced during the combustion of coal (Bahrami *et al.*, 2016). Fly ash can be used as a reinforcement for MMCs (Bahrami *et al.*, 2016; Chew *et al.*, 2011; Desai *et al.*, 2020). Desai *et al.* report on the fabrication of aluminum matrix composites with 5%–25% fly ash using stir-casting (Desai *et al.*, 2020). The authors obtained improved wear resistance, which can help improve part lifetime, as well as improvement to the hardness, yield strength, and tensile strength. Fly ash can also be produced from biomass combustion, though the properties and composition vary from coal fly ash and may vary based on the biomass combusted. Kumar *et al.* fabricated aluminum matrix composites using biomass fly ash (Kumar *et al.*, 2019). The fly ash was produced from bagasse, a waste material from sugarcane or sorghum stalks, and stir-casting was also used to fabricate the composites. The authors found increasing hardness with an increasing percentage of reinforcement material, and increases in tensile and impact strength for low levels of reinforcement (1%–3%) which decreased with greater reinforcement. The authors note that the fly ash particles themselves are brittle, and provide crack initiation points that weaken the part at higher levels of reinforcement.

Waste glass may also be used for reinforcement in MMCs. Kondoh *et al.* used glass scraps to create magnesium matrix composites (Kondoh and Luangvaranunt, 2003). Magnesium alloy based composites have good mechanical and corrosion properties at relatively light weights, making them attractive for structural or automotive components. In this work, fragments of high purity SiO₂ optical fibres ground to a fine powder were used as the reinforcement, compacted with AZ31 magnesium alloy powder, then heated to synthesis the composite in solid-state, and then hot extruded.

Additive Manufacturing of Composites and Sustainability

The respective beneficial attributes of additive manufacturing and composites, in the terms of sustainability and other benefits, have driven interest in the additive manufacturing of composite parts. A number of additive manufacturing methods have been applied to manufacture a number of composite materials. Polymer matrix composites have been fabricated using fused deposition modeling (FDM), laser powder bed fusion/selective laser sintering (LPBF/SLS), stereolithography, and laminated object manufacturing (LOM) (Yakout and Elbestawi, 2017; Carneiro *et al.*, 2015; Matsuzaki *et al.*, 2016; Shofner *et al.*, 2003; Liu *et al.*, 2018; Sandoval and Wicker, 2006; Bai *et al.*, 2013; Parandoush and Lin, 2017; Kumar *et al.*, 2012). Metal matrix composites have been produced using LPBF, binder jetting, and indirectly using FDM (Wegner *et al.*, 2020; Enrique *et al.*, 2020; Singh *et al.*, 2019; Vrancken *et al.*, 2019; Li *et al.*, 2017a). Nanocomposites have been produced using FDM, stereolithography, and LPBF (Tambrallimath *et al.*, 2019; Song *et al.*, 2019; Zhuang *et al.*, 2020).

Fused Deposition Modeling

In polymer FDM, a plastic filament is extruded through a heated nozzle onto a build plate. Relative movement between the nozzle and build plate allows the part to build up layer by layer based on the input model. For FDM fabrication of composites there are two methods: using pre-made composite filaments of polymer already containing the reinforcing fibres or nanomaterial, or supplying the matrix and reinforcing element separately and combining them in the manufacturing process.

Shofner *et al.* made composite filaments of acrylonitrile butadiene styrene (ABS) with carbon nanotube (CNT) reinforcement using Banbury mixing, compression moulding, and extrusion, then used these filaments with an unmodified standard Stratasys FDM machine (Shofner *et al.*, 2003). Carneiro *et al.* report on the successful use of a premade commercial composite filament of 30 wt% chopped glass fibre in a polypropylene matrix with FDM (Carneiro *et al.*, 2015).

Cali *et al.* also report on FDM of bio-composite filaments (Cali *et al.*, 2020). Five materials were assessed, each using a biodegradable thermoplastic PLA matrix, and an organic filler: hemp, waste powder from hemp leaf, waste from agricultural cherry tomato production, carob flour, and pruning waste from agricultural orange production. The hemp PLA composite, using 20% hemp, exhibited good tensile strength, yield strength, and stiffness, with a similar surface roughness to pure PLA or ABS, comfortable surface touch and esthetic finish, and good breathability. The authors successfully produced two biomedical device prototypes, a neck orthosis and laryngoscope, using the hemp PLA composite filament.

These methods allow convenient fabrication of reinforced polymer composites using existing FDM systems, however the effect of the reinforcement is limited for FRPs as the orientation of the fibres is uncontrolled. In FRPs, randomly oriented fibres give weaker, isotropic reinforcement, while fibres with a controlled orientation in a given direction give stronger, anisotropic reinforcement parallel to the fibres. Methods which combine the fibre and polymer matrix during the process can allow for control of the fibre orientation (Matsuzaki *et al.*, 2016; Liu *et al.*, 2018). Matsuzaki *et al.* describe such a method, where carbon fibre or jute fibre yarn are fed into a heated nozzle with polylactic acid (PLA) to deposit FRP composites (see Fig. 4) (Matsuzaki *et al.*, 2016). Jute fibre is a vegetable fibre that can be spun into yarn strands. The polymer matrix, PLA, is a plant-derived, biodegradable thermoplastic, making jute fibre PLA composites an attractive material for sustainability. The authors report modulus and strength increases of 157% and 134% for the jute fibre composites, compared to the pure PLA samples.

Liu *et al.* report on using FDM to deposit continuous carbon fibre PLA composites by combining a carbon fibre bundle with PLA filaments in a heated nozzle (Liu *et al.*, 2018). The method was applied to create strong lightweight truss structures (see Fig. 5), which are useful for aerospace applications where they can be used sandwiched between panels to provide structural strength while adding minimal weight. The authors found that the cooling extruded composite had enough stiffness to allow the structure

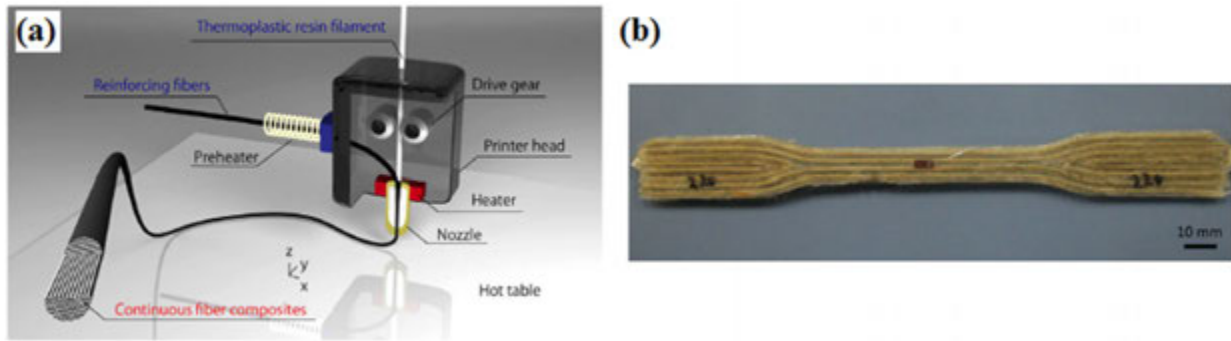


Fig. 4 (a) FDM apparatus combining carbon fibre and thermoplastic filament in the nozzle during manufacturing, (b) tensile test sample of jute fibre in a PLA matrix fabricated using the FDM apparatus. Reproduced from Matsuzaki, R., Ueda, M., Namiki, M., *et al.*, 2016. Three-dimensional printing of continuous-fiber composites by in-nozzle impregnation. *Sci. Rep.* 6 (February), 1–7.

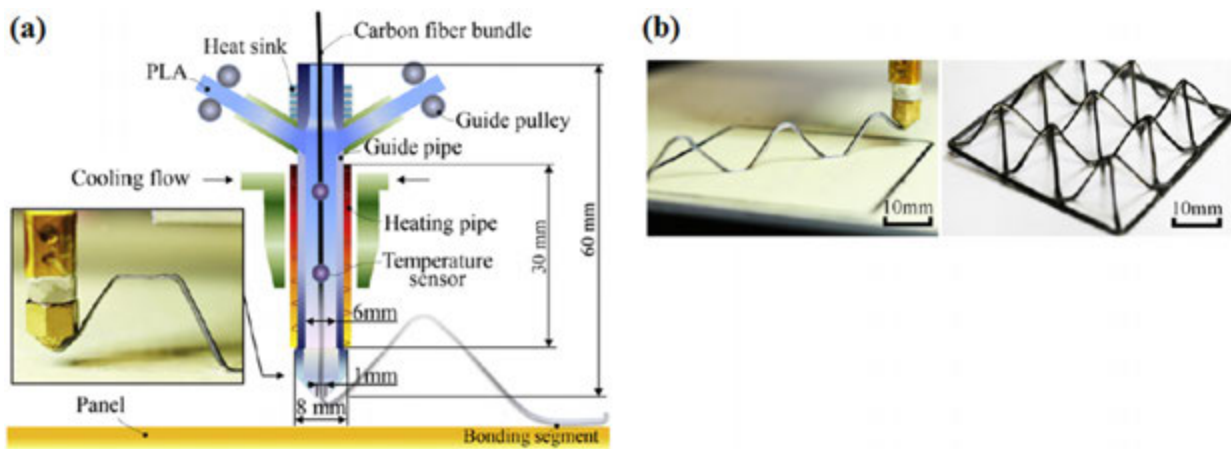


Fig. 5 (a) FDM apparatus combining carbon fibre and polymer in the nozzle during manufacturing, (b) composite truss structures manufactured by FDM for use in aerospace sandwich panels. Reproduced from Liu, S., Li, Y., Li, N., 2018. A novel free-hanging 3D printing method for continuous carbon fiber reinforced thermoplastic lattice truss core structures. *Mater. Des.* 137, 235–244.

to be printed free-hanging, without the need for support material, simplifying the process and decreasing material usage/waste. Lightweight parts aid fuel efficiency, which combined with the decrease in material use contributes to decreasing resource consumption, improving sustainability.

FDM-based industrial AM machines for production of FRP parts are available. Markforged Inc.'s x7 machine combines uncontrolled orientation fibre reinforcement with continuous controlled orientation fibres (Markforged Inc, 2020). The machine uses a composite filament of nylon, a robust polymer, with chopped carbon fibre reinforcement, providing uncontrolled orientation reinforcement. In between each layer of the build continuous fibre is wound, giving controlled orientation reinforcement parallel to the buildplate. The company report that the dual reinforced parts are stronger than aluminum 6061, while 40% lighter. This strong, lighter weight parts could improve fuel efficiencies, and thus sustainability, in automotive and aerospace applications.

Vat Photopolymerization

Vat photopolymerization is an AM method where a photopolymer resin is selectively cured layer by layer with light to make a part. Stereolithography is a vat photopolymerisation based method where the photopolymer resin is selectively cured layer-by-layer with a UV laser, and it has been implemented for the production of polymer matrix composites. Sandoval *et al.* report on stereolithography using a commercial stereolithography resin with CNTs dispersed by mechanical mixing and ultrasonication through-out (Sandoval and Wicker, 2006). The authors reported tensile and fracture strength increases of 5.7% and 26%, respectively, compared to parts made without reinforcement, which could contribute to longer part lifetimes.

Kumar *et al.* report on reinforcing a stereolithography resin with cellulose nanocrystals, also dispersed with mechanical mixing and ultrasonication (Kumar *et al.*, 2012). Cellulose nanocrystals can be derived from natural cellulose sources using hydrolysis. This renewability and biodegradability are attractive from a sustainability perspective. The nanocrystals have a fibre-like shape and high stiffness (100–143 GPa), allowing them to fulfill a similar role to carbon nanotubes or nanofibers in nano-reinforcement. The

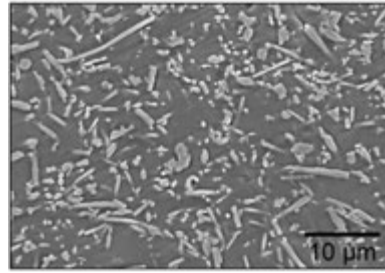


Fig. 6 SEM cross section of AM SiOC reinforced with SiC whiskers produced using digital light processing vat photopolymerisation. Reproduced from O'Masta, M.R., Stonkevitch, E., Porter, K.A., *et al.*, 2020. Additive manufacturing of polymer-derived ceramic matrix composites. *J. Am. Ceram. Soc.* 103 (12), 6712–6723.

authors found increases in the storage modulus of up to 57% in the glassy regime, and 587% in the rubbery regime, from the cellulose nanocrystal reinforcement. The percentage of reinforcement material in stereolithography may be limited if the material is opaque to UV, as the material scattering or absorbing the light will interfere with the curing of the photopolymer. Sandoval *et al.* and Kumar *et al.* successfully used up to 1 wt% and up to 5 wt% reinforcement, respectively. Zhang *et al.* produced lignin-reinforced composites using stereolithography (Zhang *et al.*, 2019). As discussed in Section “Sustainability in Composites”, lignin is an abundant polymer often available in waste materials (Haq *et al.*, 2020; Zhang *et al.*, 2019). The authors dispersed 0.2–1 wt% of softwood kraft lignin in methacrylate photopolymer resin. With just 0.4 wt% the tensile strength of the parts could be increased by 60% compared with non-reinforced parts.

Ceramic matrix composites (CMCs) have also been produced using vat photopolymerisation based methods. A slurry of photopolymer resin mixed with ceramic particles can be used to make a preceramic polymer part. These precursor parts can then be pyrolyzed to convert them to ceramic. By mixing reinforcements, such as larger ceramic pieces, into the slurry, this approach can be used to fabricate CMCs. O'Masta *et al.* report on the use of this method with digital light processing (DLP), a vat photopolymerisation AM method where UV light is projected to cure each layer, to produce SiOC matrix composites (O'Masta *et al.*, 2020). Ceramic short fibres, such as SiC, were used as a reinforcement (shown in Fig. 6) and was found to significantly improve the fracture toughness, which could improve sustainability by contributing to longer part lifetimes.

Laser-Based Additive Manufacturing

CNT reinforcement has also been applied with LPBF. In LPBF, layers of powder rolled onto a build plate from a reservoir in a powder-bed system are selectively sintered or melted to locally fuse them, building up a part layer-by-layer. Composites can be fabricated with this method by mixing of the reinforcement through-out the powder. Bai *et al.* describe the use of CNT coated polyamide 12 powders (presented in Fig. 7) to fabricate composite parts with LPBF (Bai *et al.*, 2013). The authors report increases in the flexural modulus, flexural strength, impact strength, elastic modulus, and ultimate tensile strength of 13%, 11%, 124%, 54%, and 6% due to the reinforcement. These gains were achieved with just 0.1 wt% of CNT, indicating good cost-effectiveness. CNT reinforcement can also be used for metal matrices with LPBF (Zhuang *et al.*, 2020).

Ho *et al.* report on the sintering behavior of LPBF production of graphite and polycarbonate composites (Ho *et al.*, 2002). The authors measured the graphite powder to have an emissivity of 0.81, and was highly absorbing of the CO₂ laser light, leading to increased temperatures in the powder bed. As such, graphite powder filler could improve the sinterability of powders, allowing lower laser powers to be used which gives increased energy efficiency, in addition to improving part properties. In the commercial sphere, supplier 3D Systems sells a mixed powder for fabrication of aluminum-filled polymer matrix composites under the name DuraForm[®] ProX[®] AF+ for use in LPBF (3D Systems, 2020). The company report sustainability benefits of high specific stiffness, and higher powder recyclability bringing down costs and reducing waste, improving sustainability.

LPBF is a common method for AM of metal parts, and has been used to produce metal matrix composites (Wegner *et al.*, 2020; Vrancken *et al.*, 2019; Li *et al.*, 2017a). Wegner *et al.* fabricated diamond 316L stainless steel composites using LPBF (Wegner *et al.*, 2020). The stainless steel powder was blended with 5 vol% of 8–12 μm diameter diamond powder and used to produce LPBF parts, achieving relative densities above 99%. Vrancken *et al.* fabricated Ti-Mo-TiC composites using LPBF (Vrancken *et al.*, 2019). The authors used a powder mixture of Ti with 6.5 wt% Mo, and 3.5 wt% Mo₂C to produce cubes of Ti-Mo-TiC composites using an in-house developed LPBF machine. The hardness of the composite exceeded that of AM Ti6Al4V by 150 HV. Harder, more wear resistant parts can have longer life times, improving sustainability. Wen *et al.* used LPBF to make composites with an S136 matrix and graphene oxide reinforcement (Wen *et al.*, 2019). S136 is a martensitic carbon steel with good mechanical properties and high corrosion resistance which has led to it being widely used in molds, particularly for plastic injection moulding (Wen *et al.*, 2018). Graphene oxide can be used as a reinforcement for MMCs, providing improved stiffness and strengths (Liu *et al.*, 2016; Shao *et al.*, 2018; Feng *et al.*, 2017). The authors mixed commercial S136 powder and dispersed reduced graphene oxide (RGO) sheets to coat the powder particles in 0.1–0.5 wt% of RGO. LPBF fabrication was then carried out with the coated powders. The RGO content

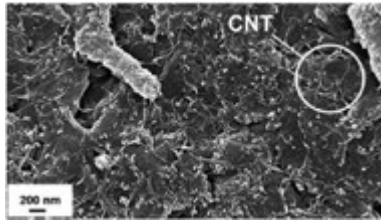


Fig. 7 SEM micrograph of the polyamide 12 CNT-coated powder. Reproduced from Bai, J., Goodridge, R., Hauge, R.J.M., Song, M., 2013. Improving the mechanical properties of laser-sintered polyamide 12 through incorporation of carbon nanotubes. *Polym. Eng. Sci.* 53 (9).

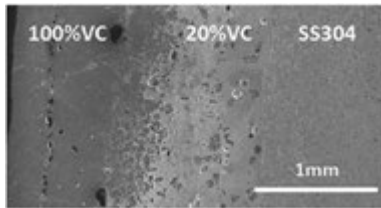


Fig. 8 Cross section of vanadium carbide (VC) 304 stainless steel (SS304) composite with a compositional gradient. Reproduced from Gualtieri, T., Bandyopadhyay, A., 2018. Additive manufacturing of compositionally gradient metal-ceramic structures: Stainless steel to vanadium carbide. *Mater. Des.* 139, 419–428.

was found to have a significant effect on the microstructure, hardness, tensile strength, and yield strength, with the best results being achieved for 0.1 wt% RGO. These property improvements could contribute to longer part lifetimes.

Laser Engineered Net Shaping (LENS) is a laser-based additive manufacturing method where metal powder is supplied directly to the focus of the laser beam (Izadi *et al.*, 2020). Gualtieri and Bandyopadhyay used LENS to produce metal-ceramic composites with gradient compositions (Gualtieri and Bandyopadhyay, 2018). Ceramic coatings may be applied to metal parts to provide a hard, refractory surface. However these coatings are at risk of cracking or delamination due to the mismatch in thermal expansion at the interface between the coating and substrate. Composites with a gradient variation from ceramic to metal, rather than a discrete boundary, may help to minimize or eliminate these issues. The authors used a LENS system where 304 stainless steel and vanadium carbide powders could be delivered by argon transport to the focus of an Nd:YAG laser beam head which can be moved in the Z-direction, above a build plate with X and Y motion. Layers could be deposited with different ratios of the powders, to produce a gradient from pure vanadium carbide to pure stainless steel (see Fig. 8). The ceramic surface was found to have an 80% lower wear rate than the substrate, and could survive harsh abrasive water jets that would easily damage the steel. This approach has the potential to improve part lifetimes, increasing sustainability.

Other AM Methods

LOM is an AM method where sheets of foil, paper, polymer, or other materials are cut into model-defined 2D layer shapes and laminated atop each other, to build up layer-by-layer into a 3D part. Parandoush *et al.* performed LOM with prepreg FRP composite tape, which had a controlled fibre orientation (Parandoush and Lin, 2017). Sheets or tapes of FRP composite are used with conventional manufacturing methods, such as lay-up or compaction moulding, giving good availability for the input material. The authors use a defocused CO₂ laser beam to thermally bond the layers of the prepreg tape (unidirectional or bidirectional glass fibres in polypropylene) together, and a focused CO₂ laser beam to cut the layers into shape (see Fig. 9). They report comparable and improved layer adhesion for unidirectional and bidirectional fibre tape, respectively, compared to compaction moulding. Lap shear strength was comparable to compaction moulding, and flexural strength was on average 30% lower, while flexural stiffness was up to 100% higher. As such, this novel method allows production of reasonably robust 3D, controlled-fibre orientation, FRP composite parts using existing prepreg input materials.

Ultrasonic Additive Manufacturing (UAM) is a method where successive layers of metal foil are mechanically welded together using 20 kHz ultrasonic transducers (Hehr and Norfolk, 2019). The oxide layers on the metals are disrupted by friction at the interface caused by the oscillation, allowing bonding between the clean metal surfaces (Monaghan *et al.*, 2015). Li *et al.* applied UAM to produce MMCs with embedded printed electronics (Li *et al.*, 2017b). An aluminum (1050) base plate was used, with aluminum (3003 H18) foils were used as the matrix material. UAM was used to create a substrate, onto which insulating material and silver-ink were deposited to create circuitry, and then UAM was used to embed the circuitry. A cross-section of the encapsulated circuit is presented in Fig. 10. With this process, the authors produced full functioning MMC structures containing embedded circuitry, with high peel resistances and weld densities. In another work, Monaghan *et al.* used UAM to incorporate metal coated optical fibres into aluminum matrices (Monaghan *et al.*, 2015). Using both aluminum and copper coated fibres, the

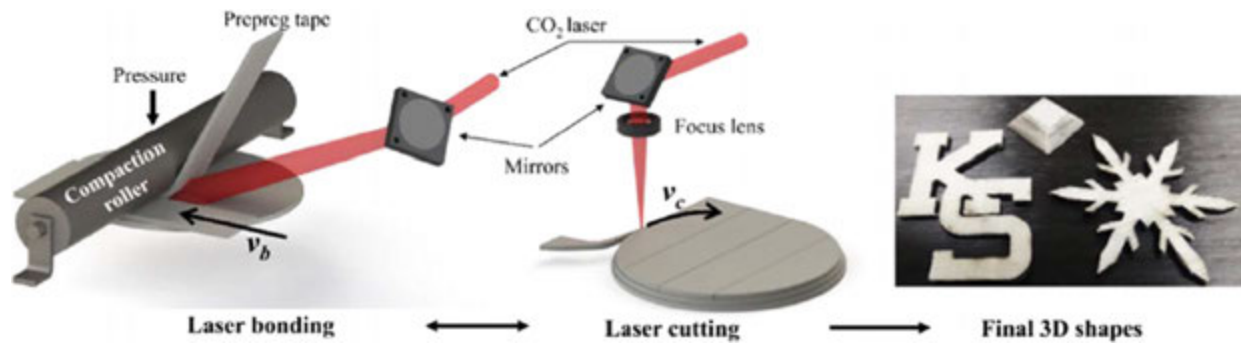


Fig. 9 LOM fabrication of parts using FRP composite tape and examples of finished parts. Reproduced from Parandoush, P., Lin, D., 2017. A review on additive manufacturing of polymer-fiber composites. *Compos. Struct.* 182, 36–53.

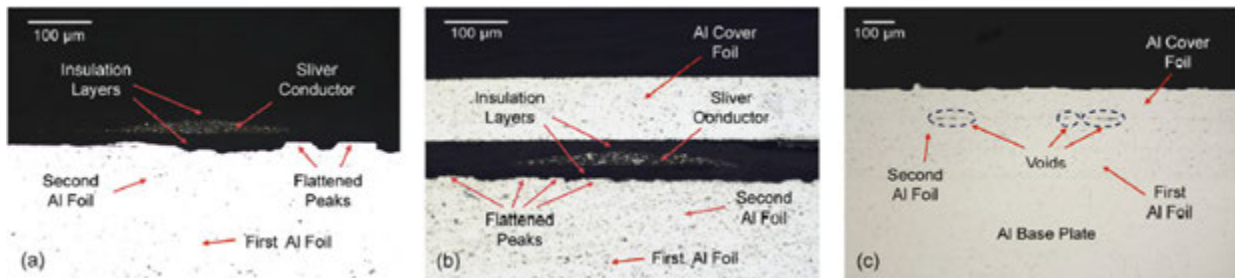


Fig. 10 Cross-section of UAM encapsulated circuitry: (a) before embedding, (b) after embedding, and (c) showing the Al-Al weld area. Reproduced from Li, J., Monaghan, T., Nguyen, T.T., *et al.*, 2017b. Multifunctional metal matrix composites with embedded printed electrical materials fabricated by ultrasonic additive manufacturing. *Compos. Part B Eng.* 113, 342–354.

authors were able to embed them in a Al 3003 H18 matrix at high energies, achieving weld densities and peel resistances approaching those of standard UAM parts.

Indirect Manufacturing With AM

Additive manufacturing may also be used indirectly in the manufacturing of composite parts. Kafara *et al.* carried out comparative life cycle assessment of mould cores, produced using conventional or additive manufacturing, for CFRPs (Kafara *et al.*, 2017). Analysing with the ReCiPe method (N. I. for P. H. and the Environment, 2011), the authors found that high impact polystyrene (HIPS) mould cores produced with AM had great potential to reduce the environmental impact. While this approach has high electrical energy usage and uses a hazardous solvent D-Limonene, it nevertheless outperformed conventional manufacturing in terms of terrestrial acidification, particulate matter formation, land usage, water depletion, and metal depletion. Continued improvement to the energy efficiency of the process and identification of alternative solvents could lead to greater sustainability improvements with AM of HIPS.

Singh *et al.* report on the production of metal matrix composites using AM assisted investment casting with recycled materials (Singh *et al.*, 2019). FDM was used to produce sacrificial patterns with recycled low density polyethylene (LDPE) reinforced with ceramic particles (SiC and Al₂O₃). The patterns were then used for rapid investment casting. When the patterns are removed from the molds, ceramic particles will be left behind such that during casting they mix with the molten metal (Al) to form metal matrix composite parts. The use of AM allows patterns to be design and made quickly, and the use of recycled LDPE is beneficial for sustainability. As noted in Section “Sustainability in Composites”, metal matrix composites may have increased strength, ductility, toughness, and wear resistance which can contribute to longer part lifetimes.

Recycled Polymer and Metal Matrix Composites via AM

As discussed in Section “Sustainability in Composites”, high specific stiffness composites are commonly used in wind turbine blades, and the recycling of the blades is a concern. Rahimizadeh *et al.* investigated the recycling of FRP composites from wind turbines to produce composite input materials for FDM (Rahimizadeh *et al.*, 2019). Short glass fibres were reclaimed from used blades by mechanical grinding, sieving, and thermal treatment to decompose the resin. The reclaimed fibres were then mixed with pellets of the relatively eco-friendly polymer, PLA, and extruded to created filaments. The authors tested use of these filaments in FDM AM, successfully producing test parts. They report specific modulus and tensile strength 18% and 19% higher, respectively, than for reinforcement with virgin glass fibres, attributing this effect to interactions between residual epoxy particles on the glass fibres and the PLA matrix.

Tian *et al.* successfully reversed the manufacturing process to disassemble AM produced continuous fibre reinforced PLA composites (Tian *et al.*, 2017). An FDM process was used, with a PLA filament being fed into a heated head along with a continuous carbon fibre, allowing both to be extruded from the head to deposit the composite. Applying a hot air gun, the polymer could be locally re-melted, allowing the carbon fibre to be pulled backwards to recover it from the part. The authors successfully reused the recovered fibre to manufacture parts with the same AM process.

Corcione *et al.* reported on the use of FDM to produce PLA matrix composites with Lecce stone scraps (Esposito Corcione *et al.*, 2018). Lecce stone is quarried as a building material, generating waste scraps in the process. Stone waste is non-biodegradable and typically disposed of in landfills (Rana *et al.*, 2016). Using these scraps for reinforcement in composites could create a useful bioplastic-based architectural material, and reduce material waste and diminish the negative effect on the environment. Corcione *et al.* carried out life cycle analysis for Lecce stone slabs with and without the contribution from landfill emissions, and identified that significant environmental improvements could be made on CO₂ emissions, ozone depletion, photochemical oxidation, acidification, eutrophication, and abiotic depletion (Esposito Corcione *et al.*, 2018). The authors produced and characterized composite filaments, and successfully used them with both an experimental and commercial FDM machine.

The powder left un-fused after LPBF production can be recycled and reused in subsequent builds, which is a good sustainability benefit, however the exposure to heat can lead to deterioration in the powders which were in close proximity to the part (Dotchev and Yusoff, 2009). This can limit the amount of powder which can be recycled without impacting the properties and quality of parts produced. Wang *et al.* report on closed-loop recycling of polyamide 12 from LPBF for use in composites (Wang *et al.*, 2018). The reclaimed unfused powder was processed into a carbon fibre polyamide 12 composite filament for use in FDM. The authors report that the parts made using the FDM filaments were comparable to those produced using commercial filaments.

For AM processes, a number of tests parts may be produced to identify suitable designs and process parameters, or to produce a number of prototypes during iterative design. This increases the material usage associated with producing a final part. Recycling of the materials from these models could reduce material waste and improve sustainability. Spoerk *et al.* assessed the recyclability of FDM polypropylene composites by pelletizing extruded material and re-extruding it (Spoerk *et al.*, 2019). A polypropylene matrix reinforced with a commercial mineral filler, FILAFORCE 96A, was used, and assessed for any degradation over up to 15 extrusions. The authors found unaltered morphology, tensile, and impact properties, and highly similar viscosities for the material after 15 extrusions.

As discussed in Section “Indirect Manufacturing With AM”, Singh *et al.* used recycled materials to produce metal matrix composites using AM assisted investment casting (Singh *et al.*, 2019). AM was used indirectly, to produce ceramic reinforced polymer patterns which when used in investment casting with aluminum created ceramic reinforced MMC parts.

Conclusion

Additive manufacturing techniques and composite materials each have their own sustainability benefits. Additive manufacturing can reduce material usage, provide lightweight parts which improve fuel efficiency, reduce transport energy usage, reduce the need for storage, and avoid obsolescence. Composite materials can provide lightweight parts with high specific strength and stiffness, improved wear-resistance and part lifetime, and in some cases can be composed of eco-friendly materials, such as biodegradable polymers like PLA, and bio-sourced additives like jute fibre or hemp. Additive manufacturing of composite materials has the potential to combine some or all of these benefits.

Additive manufacturing of composites has been successfully carried out using a number of AM methods: FDM, PBF, stereolithography, and LOM. In some cases, existing AM machines may be used without modification, but experimental and commercial systems designed specifically for composite production have also been produced. Eco-friendly composites using biodegradable polymers and bio-sourced fibres have been used with AM. Recycled materials have also been utilised in composites produced with AM, such as composites using reclaimed fibres or recycled powders from PBF. Using these techniques and materials, AM composites have the potential to improve fuel efficiencies and part lifetimes, incorporate eco-friendly and recycled materials, and reduce waste and energy use.

Acknowledgment

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Recycling of Elastomer and Polymer Matrix Composites

Vannessa Goodship, University of Warwick, Coventry, United Kingdom

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Glossary

Energy from Waste The generation of energy in the form of electricity and/or heat from the primary treatment of waste.

EU Directive Legislation passed by the European Union.

Extrusion A process technique whereby molten material is forced under pressure out of a shaped die.

Pyrolysis Decomposition brought about by high temperatures.

Gasification Decomposition brought about by high temperature to produce syngas.

Vitrimer Materials that combine the properties of both thermoset and thermoplastics materials in that they have the mechanical and thermal properties of thermosets but can be processed under the influence of heat.

Introduction

Our modern society is under an increasingly urgent pressure to reduce the environmental impact of our activities see “Relevant Websites Section”, and therefore the waste management and disposal of composite materials have also become increasingly important considerations this century, from initial material design right through to end of life (EoL). Due to the lack of ready solutions in the commercially viable space, this has been largely driven by a tightening legislative framework of environmental protection led by the European Union (EU) see “Relevant Websites section”. This developing eco-platform supports wider sustainability goals which are fundamentally based on the concepts of reduce, reuse, recycle, recover. These four “R s”, dominate our current disposal strategy. It should be noted the first of these “reduce” (waste minimization) will not be discussed within the main scope of this article, since this is a primary design issue over which there is little control by the time the materials stream reaches EoL. However, for the other options a more detailed overview of what each entail follows. This forms the regulatory platform that composite designers and engineers must work within. Countries such as Germany and the Netherlands were the early pioneers in creating challenging, commercial legislative frameworks and environments to support and seed plastic recycling activity. The automotive sector in particular was targeted to consider vehicle waste, including composites, as EU legislation set out to challenge this sector with increasingly more stringent recycling targets. ([Directive, 2000/53/EC, 2000](#)) Historically, there are therefore more automotive research programmes looking into this area than other sector groups. This Directive served as a baseline to grow and develop other legislation such as that covering the disposal of waste electronic and electrical equipment ([Directive, 2012/19/EU, 2012](#)). These directives served to initiate environmental research and development activity across both industry and academia, as well as make producers liable and responsible for the disposal of their own products.

More recently composites used in construction, wind turbines and marine sectors have become targets for improving the percentages of material that would otherwise enter landfill, and the value of striving towards a Circular Economy (CE) have been accepted. ([McDonough and Braungart, 2002](#)) Considering the decommissioning of a wind turbine as just one example, a single blade alone can be longer than the wing of an aircraft (the largest is well over 100 m in length) and offer both logistical and material disposal challenges to recycling and recovery targets.

In the past, plastic recycling research development and infrastructure was mainly utilized to deal with the high impact, high volume, low density, waste materials found in packaging applications. Therefore, whilst polymer matrix composites fall into the category of “plastics recycling”, many of the generic materials recycling systems that have been developed do not specifically cater for high end engineering composite materials. Often these materials end up as residual “other” materials losing their associated high properties and costs into low end disposal. For thermosets in particular, they were for many years considered to be “un-recyclable”, and composite recycling is still the focus of extensive research around the world. ([Overcash et al., 2017](#)).

There are a number of reasons for this contrast, not least in the design and manufacture of composite materials. With longer lifetimes in service than a packaging application, decisions about second life or disposal are already fixed and limited by the material and volumes under investigation for disposal. This waste can come in many forms, with numerous and multiple constituents, and from a number of different processes, as can be confirmed by the variety of composite materials encountered in the other articles of this book. Compared to the situation with the highly visible, more consistent, and high volume waste streams such as PET carbonated drinks bottles or HDPE milk bottles, it is apparent that composite waste streams are much less consistent in terms of their constituents, being highly tailored for specific applications, and on a much smaller scale in terms of volume in the disposal system. A further difference is the reliability and regularity of the feed stream. This is an important concept in recycling. If a material buyer wishes to design and specify a product based on recycled materials, they need to ensure there is a regular and consistent supply of material for their needs. Supply chains therefore play a very important role in the 2nd Life marketplace. ([Dertinger et al., 2020](#)).

For the purposes of this article and for ease of identifying recycling potentials, it is important to make some simple material distinctions early on in the EoL disposal decision making process. Identifying the individual polymer matrix types is of much lesser importance here, given the enormous scope of the potential matrix materials, fillers and additives that may end up in the recycling

system. The first distinction is therefore a much broader definition which relates to the possibility of melt reprocessing. Can the composite be remelted and reprocessed? This separates materials clearly into two waste streams: thermoplastic materials including the thermoplastic elastomers, and the crosslinked thermoset and elastomeric materials.

In a similar fashion, glass fiber reinforcement (GFR) recovery has received far more attention historically than carbon fiber reinforcement (CFR). However, as recycling has advanced into industrial usage and the recovery economics have been considered, the advantages in performance and economic value that recovered carbon fiber (CF) can provide over glass fiber (GF) have resulted in worldwide commercial outlets who use a variety of pyrolysis methods to recover CF and turn it into new products. Examples include see “Relevant Websites Section”. In contrast, for GF recycling, Neocomp, Germany see “Relevant Websites Section”, do undertake GF recycling, but it is not resold as fiber but ground down and mixed with other hard to recycle materials and used as an additive for cement. Recycling of the fibers in both cases is 100%, but in one case fibers are recovered for reuse as fiber (CF) and in the other repurposed (GF) to become part of an additive.

The use of thermoset versus thermoplastic materials is in itself a very interesting question for designers and the environment, when one considers that even thermoplastic materials are often coated or painted with thermoset materials to improve surface esthetics or durability.

Is a product with a longer and more durable lifetime in service, but one that is ultimately harder to dispose of, more preferential to a shorter lifetime product, but one that can be more easily recycled? For example, in the case of a wind turbine blade, there is a trade-off between being fit for purpose during the turbine blade product use phase and being recyclable at the end of the use phase (i.e. disposal/recycling).

Where durable products are required, thermoset materials can provide significant engineering solutions. However, thermoset composite manufacturers can risk their materials being overlooked entirely if environmental targets are not also aligned to consider the lifetime and durability. Equally, as our focus on the environment has changed thermosets have also gradually been replaced by thermoplastic materials (Jansen, 2020). Even thermoplastics themselves have been replaced by natural composite versions in some applications as technology develops and the environmental impact becomes of greater importance to designers. This is why it is so important to consider a whole life cycle analysis approach to composite design. We are constantly learning about our materials, processes and how best to deliver a more environmentally sustainable solution. Life Cycle Analysis (LCA), therefore, plays an increasingly important role in making the correct material choices for different kinds of application through their lifetimes.

As well as the recyclability differences between thermoplastics and thermoset materials, other general factors which must be considered at end of life include:

- (1) Quantity and volume of waste.
- (2) The reliability of that waste stream: is it a one-off waste or will it be present every year in a known volume?.
- (3) Purity (percentages of any other non-polymeric and polymeric materials if known).
- (4) Service history if known (manufacturing waste or end of life waste).
- (5) Is contamination present? and how will that impact the next stage?.
- (6) logistical considerations: does it cost more than it is worth to transport it to a recycling or final disposal facility? is there is a negative economic cost and a disposal charge to cover?.
- (7) Who will use this material?.

If some of these factors are known, then it may be possible to make some arbitrary decisions with regard to identifying the most appropriate recycling method as shown in Fig. 1. Logistics and related transport and disposal costs are a very important economic factor in sustainable recycling systems. If waste cannot be transported to recycling or recovery stations economically, then that system will have to be subsidized somewhere else in the supply chain. A further consideration is the market for the 2nd life product or the 3rd life product, e.g., can it be resold? These considerations go right through materials’ lifetimes from cradle to cradle.

The rest of this article will in Section “The Developing Scenario and Steps to Get There” consider how we can continue to develop and improve composite recycling from the current situation; Section “Reuse of Polymer Composites” discusses the challenges of reuse. Sections “Recycling of Polymer Composites” and “Recovery of Polymer Composites” detail recycling and recovery challenges and processes for composites. Section “How to Make a Composite More Recyclable?” considers what we can do to make composites more recyclable and Section “Conclusions” offers conclusions and suggests ways forward.

The Developing Scenario and Steps to get There

Returning to the questions posed in the introduction, we can consider what an ideal future scenario for composite materials in the circular economy would look like:

- (1) Composites are implicitly designed from the start to maximize their recycling or recovery at end of first life, and their LCA is well known.
- (2) Disassembly guidelines show best practice for optimum recovery of all components.
- (3) Full traceability and best practice is in place right through the system from the raw materials used, the manufacturing process, service life, and 2nd life right through to the ultimate disposal option, so the supply chain has confidence in the products they purchase and final disposal causes no environmental damage.

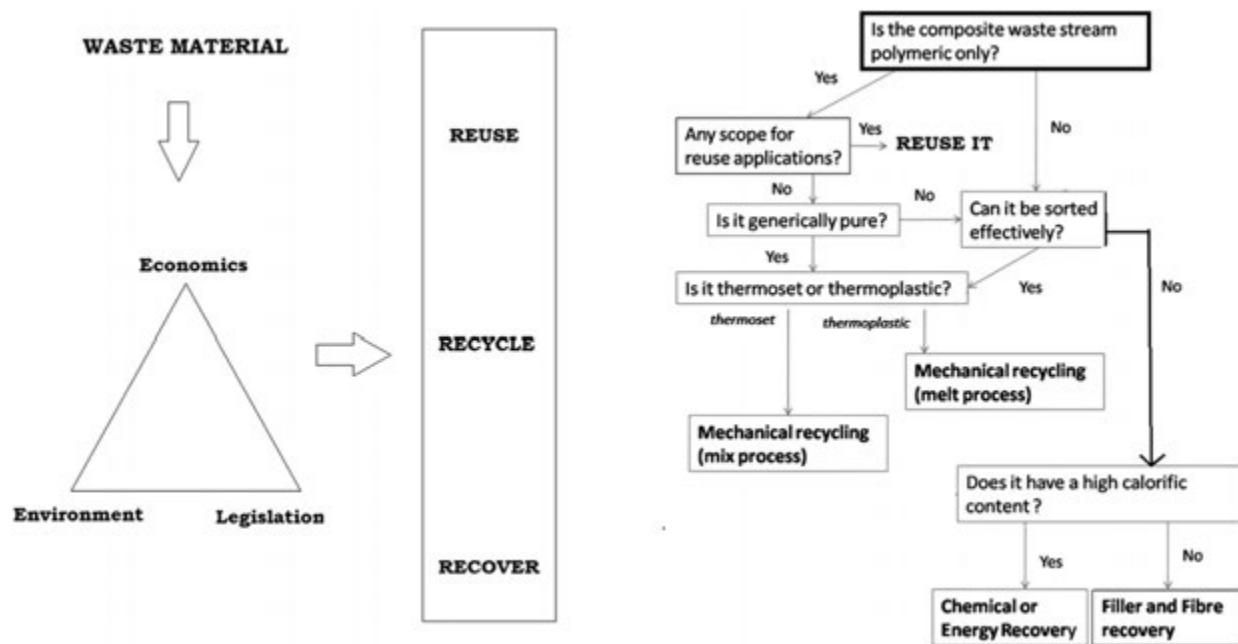


Fig. 1 Recycling options and considerations covered in this article.

- (4) Suitable recycling facilities are always sited strategically to limit transport costs.
- (5) The next use of the recovered material is always known.

In this scenario the thermoset or thermoplastic material is initially designed with the environment in mind, has minimal environmental impact and all aspects of the material composite lifecycles are optimized.

The difference between the current and the ideal environmental scenario involves a series of steps that need to be climbed. At the time of writing only the first three points can be considered to be even partially developed, and lots of work is still required to make composites more sustainable. As has already been described, one way that the European Union has sought to address this is to use environmental legislative powers to force manufacturers to deal with their waste streams. So composite raw material chemical suppliers, and the composite manufacturers industry in certain sectors must conform to stringent environmental legislation, with others likely to be subject to future legislation requirements. Similar legislation based on the 4 “R” principle already exists or is slowly developing around the world.

Reuse

Reuse means using the composite part again without further processing; examples of reuse and repurpose include using part of an old wind turbine blade to make a bench, introducing it into an architectural setting, or repurposing it for a second life whilst retaining the original composite panel component. Automobile tires have long been reused and repurposed for a variety of purposes: children swing seat, crash barriers, flowerpots, furniture etc. Many more inventive ideas can be found on the internet.

In contrast, with so many customized composite material components in the market place, this can be difficult to apply more generally for composites, and remains highly application related. A lack of performance and durability knowledge, as well as bespoke formulations and customized sizes, mean very little historical research exists in this field. However, this is slowly changing with dedicated reuse centers opening around the globe. [Reuse center, UK See “Relevant Websites Section”].

Recycling

The next step in the waste hierarchy is to recycle. Mechanical recycling of all composite materials is possible. This same process is also sometimes referred to by the term physical recycling. The available options are limited, and primarily depend on the specific polymeric component of the composite under consideration. A thermoplastic based polymer that can be melt processed generally presents few technological challenges in itself; the additives and any contaminants, however, can be problematic. Thermosets have to be ground down and used as part of another formulation. The difficulties tend to be attached to costs: collection (logistic costs) and recycle preparation such as shredding and grinding, separation of other materials (sorting) and its associated technology (Makenji, 2010). Contamination mitigation (Al-Salem *et al.*, 2009), labeling and traceability, and logistics (Mariano *et al.*, 2017) are covered elsewhere and beyond the scope of this article which has a materials focus. Research has been undertaken on the mechanical recycling of various polymers and additives, and across sectors; however, there still needs to be a market for the

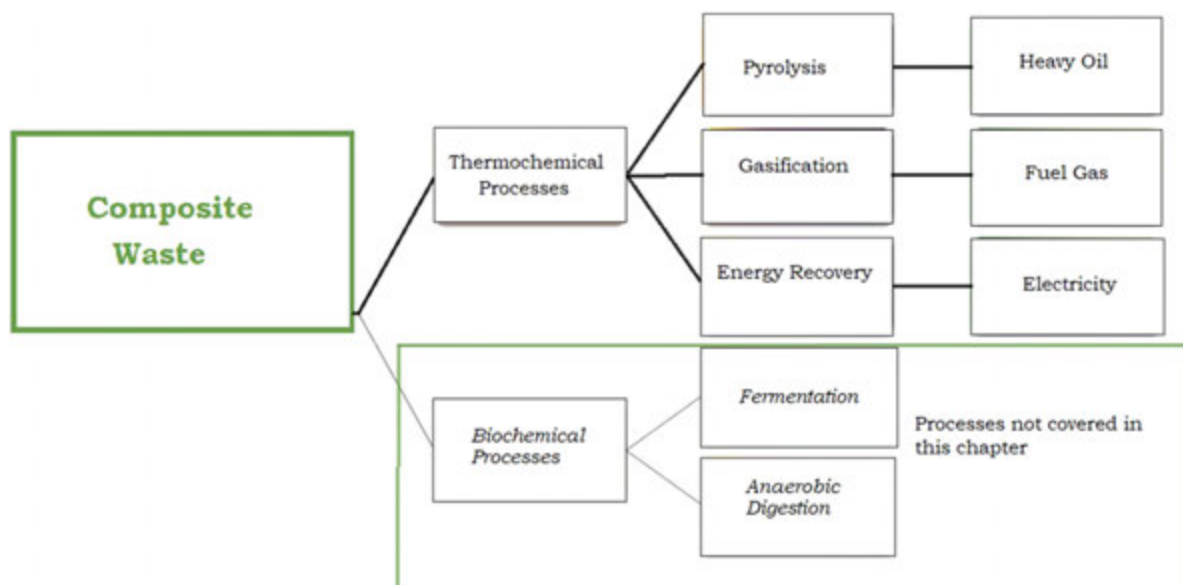


Fig. 2 Processes for recovery (biomass, energy and materials).

recycle material produced, which can be another barrier for composite 2nd life applications. Beyond mechanical recycling, thermal methods are used for further recovery of constituents and/or energy recovery.

Recovery

Rather than mechanical methods, the term “recovery” here is used to cover all thermal and chemical breakdown methods. This can be anything from recovery into constituent chemicals for creating new polymers (feedstock recycling), fiber recovery or simple incineration of waste materials with or without heat recovery, though obviously heat recovery is preferred in the waste management hierarchy. As in mechanical recycling, there is often a need to first size reduce the size by mechanical means.

Once this initial step is complete, further processing can be split into the following options:

- (1) Thermal treatment to obtain partial material recovery by pyrolysis and fluidized bed techniques.
- (2) Chemical recovery to obtain conversion of waste material into chemicals and feedstocks.
- (3) Energy recovery. The calorific value of the component can vary greatly dependent upon the percentage of polymer, additives and matrix materials as well as purity of feedstocks. This factor can greatly influence the viability of different routes for recovery for a composite materials.

It should be noted that biomass recovery also falls under this category; this is of relevance to the growing field of bio-composites, which is not covered here.

A quick word on landfilling. When the European Directive 1999/31/EC on the Landfill of Waste (Directive, 1999/31/EC, 1999) came into force, it was with the implicit intention to both minimize material going into landfill and ensure that any waste which was landfilled did not harm the environment. This is the least preferred waste management option for disposal.

A volume reduced waste stream from the reprocessing of composite waste can be residues from other treatment processes such as incineration in the form of fly ash or bottom ash. This can still contain harmful material residues depending on the original composite formulation. The disposal options of these composite residues may be different from one country to the next. (Sauve and Van Acker, 2020) An overview of available recovery and disposal systems is given in Fig. 2. provide good coverage of the environmental impact of landfills.

Reuse of Polymer Composites

Composite waste is produced from a variety of industrial markets. Considering just the wind energy sector alone it is estimated that 2.5 million tonnes of composite materials are currently in use on wind turbines which will require decommissioning within the next 20 years. see “Relevant Websites Section”. For sectors such as wind energy, which are heavily reliant on thermoset plastics, this presents a significant recycling challenge, and therefore a full range of viable disposal options will need to be considered within the local legislative framework in which they are used. Reuse of large thermoset structures such as a decommissioned wind turbine blade are being actively researched on projects such as Re-Wind [<https://www.re-wind.info/>], and designed reuse of large sized pieces have been reported (Bank et al., 2019). It has been proposed that where harsh environmental conditions exist, a piece

of a wind turbine blade could create a durable construction material. In Mexico, for affordable housing projects, usage has been proposed for both structural and architectural applications (Bank *et al.*, 2018). Other potential reuse/recycling issues focused on construction are described by Conroy *et al.* (2006) and Yazdanbakhsh and Bank (2014). Other potential reuse/recycling issues that focus on construction are described by Conroy *et al.* (2006) and Yazdanbakhsh and Bank (2014). According to their research, the lack of durability knowledge of composites used in this way, as well as bespoke formulations, generally makes reuse applications in construction rare, although projects may become more common as interest in reuse and upcycling grows.

Limiting complexities arise in reuse of composites as a result of the various fixtures and fittings used in each specific application. Whether it is honeycomb core material, metal inserts used as fixings or reinforcement, various drilling and fixing sites in the composite all add to the bespoke nature, design complexity and limitations of reuse.

Major problems for composite reuse can therefore be summed up as:

- (1) Bespoke designs.
- (2) Mixed materials (composite, foam, metal).
- (3) Relatively small disposal volumes.
- (4) No reuse applications or market.
- (5) The logistics and cost of decommissioning: This is a significant on cost if for example considering the decommissioning of a wind turbine farm.
- (6) Confidence in the structural integrity of a used composite item: This is likely to remain a large ongoing problem for this market.
- (7) Limited data associated with composite materials as second-hand parts and upcycling on the marketplace.

Recycling of Polymer Composites

At a lower level of the waste management hierarchy are recycling and recovery.

Grinding up a composite or elastomer and using it as an additive/component of another material, such as the example of the GFR composite used as an additive in concrete materials already discussed, has been extensively reported (Siddiqui *et al.*, 2008) and is carried out commercially. For example Global Fiberglass Solutions see “Relevant Websites Section” recover and shred wind turbine blades for their products thereby diverting them from USA landfilling sites.

All composites including vulcanized tires and elastomers can be ground down and potentially used as filler materials into other plastic composites. However, this can be detrimental to the mechanical properties of the resultant composite if not carried out optimally. It should also be considered that while simply grinding down a high performance material as a cheap filler does satisfy the requirements of disposal, it is a significant downgrading of the properties of the original material.

Mechanical Recycling of Polymeric Matrix Composites

Preparation of recyclate

Mechanical recycling requires materials to be first prepared for reprocessing. This can involve a number of stages such as size reduction, sorting and cleaning. These stages are necessary to reduce the quantities of any non-targeted materials and/or metals and glass. It is also necessary to remove contaminants such as oils, glue residues, dirt or others.

A typical thermoplastics recycling plant may include a number of processes as shown in Table 1.

A more recently reported sorting technology for composite components is electrodynamic fragmentation. This underwater process works by firing ultrashort (< 500 ns) discharge pulses, which travel along the composite phase boundaries and break the component into smaller pieces. This has been demonstrated on CF filled polyether ether ketone (PEEK) thermoplastics by (Roux *et al.*, 2015), who were able to reuse the fragments; however, there was a reported 17% decrease in mechanical properties due to fiber length reduction.

Table 1 Typical processes for thermoplastic composite recycling

Process	Variants
Size reduction	Uses equipment such as bale openers and shredders to reduce components.
Separation	Drum screens. Vibrating screens Gravity/density separation such as float - sink, hydrocyclones, air classifiers, ballistic separators. Magnetic and eddy current separation for metal separation: permanent magnetic separators or electromagnetic separators Optic sensor separation covers processes such as NIR Infrared , an in-line camera or X-Ray Fluorescence
Washing	Remove contaminants with water, detergents and agitation. May also include removal of glues.
Dryers	To pre-dry material prior to extrusion.
Extruders	Melt screens: during extrusion the plastic passes through a screen in order to remove any solid impurities. Pelletizing to produce granules.

Extrusion

Once material is prepared it can be extruded alone or in combination with other materials and additives. For extrusion reprocessing material is required to flow under pressure, therefore fully cured materials are not suitable unless being used as a minor component as mentioned in the introduction to this section. Therefore the inherent chemical nature of the polymer matrix: thermoset or thermoplastic, rather than the additive components will often dictate the reprocessing options available. This section is now split into considerations for thermoplastic and then thermoset mechanical recycling. Elastomers should also be considered as either (1) being able to melt processed or (2) fully cured in this respect. It should be noted that a common issue exists for all materials in the retention of properties following this process, specifically for the key composite property of fiber length retention, this must be considered.

Mechanical recycling of thermoplastic matrix composites

The drive for lightweighting and the circular economy has seen renewed interest in the use of thermoplastic matrix composites in replacing both metal and thermoset materials in some applications where technically feasible. In house recycling of waste in thermoplastic manufacture and production is considered standard good practice so often these kinds of wastes are absorbed by the factories that produce them. This is because thermoplastics can be made to flow and harden repeatedly through a cycle of action of heat, pressure and cooling, in contrast to thermoset manufacturing waste. Once they reach end of life however, they become part of the waste stream. Generic recycling of thermoplastic materials has been widely studied for many years especially for common matrix polymers such as PE, PP (Tapper *et al.*, 2018) (Jansson *et al.*, 2003), HDPE, PA (Eriksson *et al.*, 1997) and PET (Awaja and Pavel, 2005); thermoplastic elastomers can be reprocessed by similar methods (Fazli and Rodrigue, 2020). Recycling data for more high performance reinforced matrix types such as PEEK (Roux *et al.*, 2015) is also available.

Due to mechanical shearing processes present in both grinding and extrusion, long fiber lengths are reduced during reprocessing. This has been known for many years, and without exception is the case for all polymers which show the same trends in fiber reduction. (i.e., PET (Cornier-Ríos *et al.*, 2007; Blazó, 2010) (Bernasconi *et al.*, 2007) and PBT (Chu and Sullivan, 1996)). Equally, the polymer composite and the additive interfaces can be subject to degradation mechanisms caused by the pressure, heat and shear of reprocessing. Given the large variability of formulations available, different composite processing routes and any effects on resultant interfacial bonding, separating the inter-relationships between the matrix and reinforcement and/or additives is a complex problem. However, with the clear link between mechanical properties to fiber length, it is virtually impossible to retain original properties under any form of mechanical reprocessing step with thermoplastic composites with long fibers. An example of a possible solution would be reshaping by a process such as stamp forming, whereby a sheet composite could be repressed to avoid the need for shredding and extrusion stages (and fiber reduction). However, this is not currently represented in the literature.

So while mechanical recycling effects can be understood on a general level, there is still a requirement to understand the effects of the reprocessing parameters on particular formulations. Studies in PET- GF recycling (Qui *et al.*, 1999) suggest a highly complex interaction with properties such as the fiber volume fraction, diameter and length, orientation, distribution and the interfacial adhesion between the glass and the polymer all effecting outcomes. Add to that the changes in the polymeric component, other additives in the polymer and effects on subsequent morphology, and a highly complex mixture emerges.

Therefore, any differences in polymer matrix, the shape of glass and its loading, and other additives present make individual comparisons of results problematic, and that is even before differences in the parameters of materials preparation and reprocessing. There are also suggestions in the literature that the size of the original particle is a key factor in whether it will be subject to shear breakages. This may have implications for nanocomposites (they will be mentioned later). For example, in a well detailed study by (Lu and Malloy, 1999) on Nylon 6, the inclusion of talc with glass fiber has been shown to offset property loss caused by changes in fiber aspect ratios. There may be many such interactions occurring.

Our current mechanical recycling efforts continue to be driven by more visible and voluminous waste streams such as packaging plastics, with lesser focus on specific issues affecting composites.

There is no doubt mechanical recycling alters the relationships between the inherent physical and chemical interactions and resulting behavior. We can speculate, but currently we are not entirely certain where thermal or mechanical damage (or even some property enhancement) is occurring for these systems, unless more individual formulations are studied with the same set of parameters to allow a systematic detailed comparison.

The topic of recycling of nanocomposites on both a formulation and environmental infrastructure level (Sánchez *et al.*, 2014) could be a significant factor for a future composite market if nanomaterials such as tubes, fibers or whiskers are used more widely. The effects of the incorporation of nanoparticles on thermoplastics was studied by (Sánchez *et al.*, 2014); they concluded that mechanical recycling processes would have to be adapted as nanoparticulates became more prevalent.

Given the issue with pure materials, it becomes even more complex when there is contamination present. Maris *et al.* (2018), provide coverage of compatibilization strategies for such mixed plastic waste (MPW) systems for common waste polymers. However, compatibilization adds a further process cost, which can impact the commercial viability of recycle.

Due to mechanical shearing processes present in both grinding and extrusion, long fiber lengths are reduced during reprocessing. This has been known for many years, and is the case for all polymers which all show the same trends in fiber reduction. It should also be considered there will be a further stage of potential damage when subsequent extrudate is sold on for conversion or manufacture by processes such as extrusion, compression molding, injection molding or blow molding.

Biodegradable composites have not been covered here as they would be directed into separate waste infrastructure; however, it should be stated that environmentally degradable biomaterials in large quantities can contaminate mechanical recycling routes. It

is worth noting that the fiber reduction effect of mechanical reprocessing is also present in natural fibers such as kenaf, sisal, hemp, and wood fiber.

Mechanical recycling of thermoset matrix composites

Given that considerable mechanical damage does occur to the fiber length during grinding and granulation stages, once cured, the only way to reduce the size of a cross-linked material (thermoset or thermoset elastomer) is to physically reduce it. As a result the comminution (mechanical pulverizing and shredding to reduce particle sizes) of waste is necessary to recycle these materials. This is achieved by any number of mechanical means (saw, shred, impacting hammer etc.) followed by sieving into sized fractions as required. This results in slightly different size reduced outputs varying between fine powder to fibrous products. There is not a great deal of literature with regard to mechanical recycling as other process routes are generally preferred. In terms of mechanical recycling after size reduction, this can be split into direct use in formulations that are either thermoplastics (as a powder filler), or for use in formulations that are thermoset.

It should be noted here that fiber fractions are also recovered from thermoset composites using processes found in the recovery section of this article. The polymeric components in those cases are used as fuel. These fiber only fractions, can be put into new formulations and produce mechanical strengths in the region of 80%–90% of the original fibers. There are considerable economic benefits to doing this for an expensive fiber such as CF as represented by the number of commercial operations for CF. In contrast, it is uneconomical to do this for GF; hence GF wastes are directed to other outlets such as concrete.

Beauson *et al.* (2016), used shredded polyester GF composites from wind turbine blades to make new polyester composites, but found the resultant mechanical properties to be poor. It has also been found to be detrimental to retained mechanical properties by Derosa *et al.* (2005), using Sheet Material Compound (SMC) into Bulk Molding Compound (BMC) and Silva *et al.* (2012) who also looked at polyester GF recycling and found significant reductions in properties of the new composites.

Kouparitsas *et al.* (2002), took a slightly different approach by mechanically recovering a highly fibrous fraction from a thermoset, but then reusing it in a thermoplastic (PP-GF) rather than a thermoset material. As the authors report, properties were mostly retained in the thermoplastic, but this author's opinion this is likely to be due to the significant difference in the original comparable polymer fraction properties rather than any significant retention effect. None the less, a clear potential recycling use path is shown.

Thermoset elastomers have been used as both a filler and a matrix. Ansarifar *et al.* (2010), looked at using GFR plastic waste as a fine powder filler in styrene-butadiene rubber and was able to improve some properties such as tensile strength, tensile modulus, and elongation at break, as the loading increased. In contrast, Aoudia *et al.* (2017) used microwaves to assist devulcanization of tire rubber. This was subsequently ground and fed as a filler into new epoxy composite materials to improve properties.

From studies such as these and others (e.g., Palmer *et al.*, 2009), it has been found that it is necessary to prepare and sieve specific fractions and then to optimize subsequent formulations and properties. The economic costs and therefore commercial viability to do this, mean mechanical recycling of thermoset material is not currently an attractive method of disposal.

As in the mechanical recycling of thermoplastics, the reduction in key mechanical properties such as strength and modulus can often be attributed to fiber length reduction. Similarly, variations in reported processing methods make comparisons across different studies difficult. It also needs to be factored in that the recovered glass retains residue of resin, coupling agents and any other fillers present near the interface. This produces a partial glass fraction rather than 100% GF fraction for reuse which will subsequently affect resultant properties such as the adhesion, mixing and wetting in any new composite material. It is therefore not surprising that the mechanical performance is somewhat reduced.

There are several ways to mitigate performance reduction: select fibers with longer retained lengths, increase the percentage of fractions to incorporate similar reinforcement levels, as well as to consider processing needs to aid wetting out. Together, it may be possible to get nearer to the levels of similar virgin formulations. As referenced previously, this has also been found when applying similar methods to placing thermoset material into thermoplastics. Where the fibers are used in a targeted way, and the inherent properties of the recycle considered, optimized results can be produced. This will need to be done if mechanical recycling of thermosets is to find more viable outlets for the future.

Recovery of Polymer Composites

A completely alternative approach to a mechanical recycling is to break down the composite and try to recover the component chemical parts and/or the inherent energy within. Some of these processes are undertaken on a commercial basis. Both Carbon Conversion, USA and ELG Carbon Fiber Ltd. in the United Kingdom use undisclosed, commercially sensitive types of pyrolysis technologies to recover and resell the CF fraction of thermoset waste in their products. Recovery methods can be thermally driven reactions like pyrolysis or chemical driven reactions; this section will be split in this way.

Thermal Conversion Methods

Thermal conversion methods for plastic recycling have been extensively researched and accepted as a means to managing plastic waste. Particularly for thermosetting polymers, with the difficulties in viable mechanical recycling, separation and recovery of the

phases are necessary in order for components of these materials to be widely reused. The fundamental differences between thermoset and thermoplastic properties are generally not important to thermal processes. Both pyrolysis and solvolysis have been shown to produce reusable fibers for highly reinforced composites with retained properties in the order of 80%–90% of virgin stock. Given the many variables, this article has not quoted actual values. Research from 2012, reported “recycled material maintains between 65% and 94% of its static properties and 94% of its specific energy absorption capability”. (Meredith *et al.*, 2012) Given that there are many factors at play in considering one composite formulation versus another such as comparing polymers, fiber lengths, fiber aspect ratios, and interfacial adhesion, it would not be appropriate to offer a single value here. However, developments in recovery processes and therefore the retained strengths of fibers are rapidly developing and widely reported in the scientific literature as well as commercially available specifications.

A range of different methods and processes will now be introduced, beginning with the simplest; controlled burning of waste.

Incineration and energy recovery

Combustion is an exothermic chemical reaction of a fuel with oxygen. Common products of this reaction are water and carbon dioxide with the release of heat energy. Simple direct combustion systems work by oxidizing viable solid waste to produce hot flue gas. This can be used for either a direct heating system or as a boiler to generate turbine steam for electricity. As polymers are made from petroleum (or other similar equivalent natural oils), they are a good fuel source.

Combustion of filled composites can also be undertaken to produce a waste volume reduction, but only if the polymer matrix materials and other combustibles are there in sufficient quantities to generate thermal energy. Any residues that result from non-combustible components would still require a further disposal process. These residues are commonly termed fly ash and bottom ash. Flue gases can include water vapor and carbon dioxide and these and other generated gases can be used for heat generation. For energy from waste plants (EfW) with coupled energy recovery processes, this energy is generated and collected. A key consideration for this process is the level of organics in a composite waste stream. Calorific values for major thermoset resins, as well as the negative impacts of fillers and additives related to combustion processes, can be found with reference to (Pickering, 2006). Ash residues disposal methods are to use in cement (Clark *et al.*, 2020), in other plastic products (by mechanical recycling), or where still permitted, into landfill sites.

As the calorific value varies depending upon the loadings, this can have a significant impact on the suitability of the process. For example, a polyester (calorific value of 30,000 kJ/kg⁻¹) has a substantially reduced value at 50% glass loading (15,000 kJ/kg⁻¹). Some additives also absorb energy when combusted, for example, the common filler calcium carbonate. This absorbs 1800 kJ/kg⁻¹ when it is decomposed at temperatures in the range 700–900°C. SMC has a low calorific value of just 6680 kJ/kg⁻¹ which makes incineration an unsuitable disposal option. Therefore, further thermal processes have been explored for highly loaded FRP, examples are processes such as pyrolysis and gasification.

Thermal recycling techniques

Rather than just recover heat, thermal treatment processes such as pyrolysis and gasification can be used to recover the solid, liquid and gas fractions. These can be used as feedstocks or fuels for other processes. These cracking processes can be used to separate the organic and inorganic matter and overcome any issue related to processes based on calorific values alone, as found in the energy recovery process. A commercial example of this is see “Relevant Websites Section”, who market the Thermolyzer™. This is a modular waste-to-energy reactor system, which can process materials including cured and uncured waste, vulcanized tires, as well as recover by-products (e.g. a trademarked natural gas replacement product which is used to power the process itself, with residual char and emitted exhaust gas as by products).

Thermal recycling can involve multiple process steps and chemical agents. Typical steps are ordered as follows: (1) heat and dry the material; (2) carry out pyrolysis to drive off the volatile fractions (no oxygen present); (3) oxidation processes (oxygen present) with (4) gasification of the remaining charcoal. An advanced discussion on how these processes work is beyond the scope of this article; however, a brief description will be given along with relevant composite research.

Pyrolysis

Pyrolysis can be defined as the thermal degradation of organic materials. This can be done in a variety of environments: in the absence of oxygen, with oxygen or in steam. It results in the production of solid (charcoal), liquid (oil) and gas fuel products. (i.e., CO, CO₂, H₂, CH₄, C₂H₆). It can be used to separate organic and inorganic fractions, by decomposing the polymer matrix down to chemicals which can be evaporated off. The reinforcement can also be recovered as a solid residue. More specific details of the temperatures and products of pyrolysis in relation to specific polymeric components can be found in the work of (Blazó, 2010). A variation on this process is microwave pyrolysis which enables faster energy transfer and resultant energy savings (Åkesson *et al.*, 2012).

Pyrolysis chemical reactions are strongly influenced by both the process parameters being used and the type of feedstock being processed. Reactor design and operating parameters are therefore important. Pyrolysis processes can be further categorized according to the material conversion rates. Examples are fixed-bed reactors (slow pyrolysis), fluidized bed reactors (fast/flash pyrolysis), rotary kilns and screw pyrolysers. Common parameters include temperature, heating rate, mass feeding rate, feed particle size, and residence time in the reactor. These can all affect the reactions taking place. Some example polymers, temperatures and resultant products are given in Table 2.

Liquid products are generally formed at lower temperatures than the gaseous products.

Table 2 Example polymer behavior in pyrolysis

Feedstock		Process temperatures	Products		
Polymer	Atomic ratio H/C	Pyrolysis (°C)	Liquid	Solid char (%) ^a	Residual mass (% wt) ^a
Epoxy	1.1	370–460	Alkylphenol	15	15
PA6	1.8	430–490	Aliphatic	0	0
PBT	1.0	370–430	Aromatic acid/aliphatic	3	3
PC	0.9	480–570	Alkylphenol	22	22
PET	0.8	400–460	Aromatic ester	11	11
Polyester	0.9	370–460	Aromatic	26	26

^aVaries with process conditions.

Note: Blazó, M., 2010. Chapter 5 – Pyrolysis for recycling waste composites. In: Goodship V. (Ed.), Management, Recycling and Reuse of Waste Composites, Woodhead Publishing Limited, 102, ISBN 978-1-4398-0104-8.

Table 3 Recovery of a polyester matrix composite by pyrolysis

Pyrolysis process target	Conditions
Liquid and gas fractions	Carried out in steam atmosphere at 600 and 700°C. Pyrolysis oil was high in styrene. The gas produced was mainly CO ₂ .
Glass fibers	Steam atmosphere at an optimal temperature of 700°C.
Separation of the glass fibers from CaCO ₃	Calcium salts were dissolved in hydrochloric acid.
Removal of residues on the surface of the fibers	Burnt off at 500°C
Explore highest operating temperature	900°C: Caused degradation of the glass fibers in the presence of CaO.
Optimal process parameters (lowest effort for recovering glass fibers): 600 and 700°C in steam atmosphere (with acceptable small losses in organic products).	

Note: Adapted from Grause, G., Mochizuki, T., Kameda, T., *et al.*, 2013. Recovery of glass fibers from glass fiber reinforced plastics by pyrolysis. Journal of Material Cycles and Waste Management 15, 122–128. Available at: <https://doi.org/10.1007/s10163-012-0101-x>.

Organic pyrolysis reactions generally occur at the lowest operating temperatures of 400–700°C but can be as low as 250°C and as high as 800°C; above 800°C carbonization occurs. Gasification (discussed in the next section) takes place at higher temperatures in the range of 800–1100°C.

As well as recovering polymer components as chemicals, it is also necessary to recover other fillers and reinforcements. Common additives such as calcium carbonate, glass material and carbon material recovery have all been successfully achieved (Meyer *et al.*, 2009). Unfortunately, it is not always possible to optimize recovery for all components of a composite as recovery conditions may be different. For example, char residues can leave unwanted coatings on recovered fillers and fibers which subsequently need to be removed. Char formation is related to the atomic ratio of Hydrogen (H) and Carbon (C), and Char will form readily at ratios below 1.

Grause *et al.* (2013), studied the pyrolysis of a polyester resin composite material. It consisted of an unsaturated polyester resin, cross-linked with styrene, and contained both GF and CaCO₃. The composite recovery methods are shown in Table 3.

More recent research into pyrolysis has looked at waste electronic composites disposal, for example, such as for disposal of brominated epoxy printed circuit boards (Ma *et al.*, 2020). Brominated flame retardants (BFR) are toxic and subject to legislation (Directive, 2012/19/EU, 2012) (Directive, 2011/65/EU, 2011). The European Union has set limits on the maximum concentration of BFR in electrical and electronic equipment to below 1000 ppm. Since BFR are present in composite materials, this also impacts on the recycling and recovery of these materials. Low cost commercial methods to extract the Hydrogen Bromine from the resultant pyrolysis oil from waste printed circuit boards are being investigated.

Other strategies for improving process energy or product recovery in pyrolysis reactors include investigations into vacuum pyrolysis, microwave assisted pyrolysis and catalytic pyrolysis.

Waste tire pyrolysis can also be undertaken, to produce liquid oil or tire pyrolysis oil. Tire char can be reutilized as carbon black or upgraded to activated carbon. Yields of gases are lower than the oil and char, and therefore the gas tends to be fed back to provide energy for the process (Martínez *et al.*, 2013).

In also considering the economics of commercial systems for thermoset materials, (Gouet *et al.*, 2020), developed a two-stage catalysis pyrolysis method to produce a saleable product, carbon nanotubes, from phenolic formaldehyde resin. These kinds of developments will allow the valorization of waste to make a significant contribution to the CE.

Gasification technology

Following pyrolysis with resultant liquefaction to obtain oil products, gasification is a further thermochemical conversion process to convert carbon or hydrogen containing substances into gas by use of a gasifying agent such as air, steam or oxygen. The gas product is commonly referred to as syngas, and consists of simple useful gases such as hydrogen (H₂), carbon dioxide (CO₂),

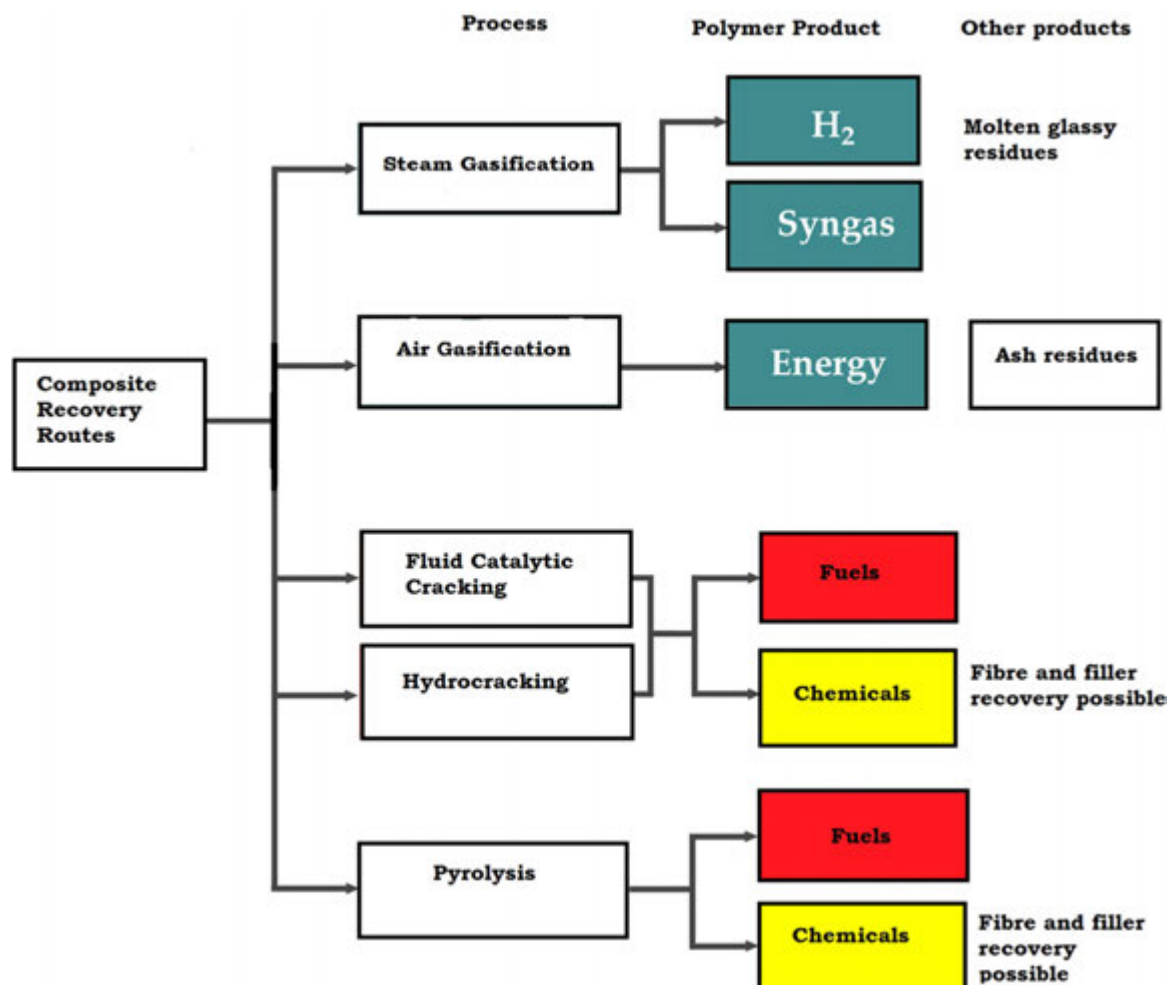


Fig. 3 Thermal recovery process routes and products.

carbon monoxide (CO), water (H₂O), and fractional quantities of hydrocarbons such as methane (CH₄). An overview for plastics waste fractions can be found in the work of [Lopez et al. \(2018\)](#).

Gasification is less sensitive to material mixtures compared to pyrolysis, which makes it more suitable for mixed polymeric or elastomeric feedstocks which can be valorized. This process has also been developed to also enable CF recovery, but as gas can only be generated from organics, highly inorganic loadings of composite materials are not ideal. However inorganic loading can still be tolerated by this process. This is an oxygen deficient process and the generated syngas can be used directly as a fuel, or following catalytic or cleaning conversion processes (to prevent the formation of dioxins and furans) it can be converted to liquid fuel or chemical intermediates.

The ash produced from high-temperature gasification is still molten and once quenched can be used as fillers in cements. Although some commercial gasifiers can recover molten metals, they do not currently recover the composite reinforcements. Further details on gasification research can be found in [Lazzarotto et al. \(2020\)](#).

A summary of thermal recovery routes can be found in [Fig. 3](#).

Depolymerization Technologies

The next sections are concerned with chemically rather than thermally breaking down the polymer. An example is Solvolysis, whereby the resin is dissolved within a solvent system, to leave the reinforcement.

Solvolysis

This is a generic process term for chemical reactions (the name is based on the word solvent). It therefore includes processes such as hydrolysis (solvent: water), methanolysis (solvent: methanol) and glycolysis (solvent: ethylene glycol). It also covers the use of the subcritical and supercritical solvents. Solvolysis therefore offers a wide range of potential routes by using different solvents, temperatures, pressures and catalysts. ([Oliveux et al., 2015](#)).

One advantage of this route over pyrolysis is there is no residual contamination by char on the fibers, enabling better reuse properties for subsequent recycled fibers. However, few commercial outlets for any these technologies appear to be operating at present. see “Relevant Websites Section” offer the process of a wet chemical treatment rather than operating a plant itself. The process uses a solvent and catalyst and is claimed to operate with low heat and pressure; however, no temperature or pressure guide was detailed.

Whilst hydrolysis reactions are the most explored method in academia, current activity in chemical methods include those that are based on supercritical solvents such as supercritical water (Kim *et al.*, 2019). Be it with water or an organic solvent such as acetone (Okajima and Sako, 2019), with the presence of a catalyst or without one, the supercritical solvents are heated ($> 150^{\circ}\text{C}$) and operated under high pressure. (Zhao *et al.*, 2019), used a microwave assisted catalytic method to chemically recycle epoxy. It should be noted that trend to integrate microwave technology was also explored in pyrolysis to aid energy recovery. Energy consumption of such processes is high, so there still needs to be further development of methods that can operate at reduced temperatures and pressures to pursue the green design principles proposed by Anastas and Zimmerman (2003). In terms of comparisons of environmental impacts, (Khalil, 2018), used LCA to compare the environmental and human health impacts for both pyrolysis and solvolysis (using SCW) of CF filled polymers. It was found that currently recycling via pyrolysis is more advantageous to both the environment and our human health.

Acid digestion

Chemically decomposing waste composites can also be achieved using acid digestion. This can avoid the use of high temperature processes to recover inorganic components. Acid dissolution of both SMC composites and amine-epoxy composites has enabled fiber recovery in laboratory studies (Ma *et al.*, 2017). However, currently there appears to be no obvious advantages to this method over others as there is a requirement to use hazardous chemicals to dissolve the composite components. For example, in order to dissolve the calcium carbonate in SMC orthophosphoric acid is used, for epoxy-CF composites, oxidative peroxide digestion has been reported to recover near virgin quality carbon fiber. However, the process is not currently viable to scale-up as the solvent is both costly and explosive, so alternative acids are needed.

How to Make a Composite More Recyclable?

The current and future technologies for reuse, recycling and recovery methods for composites and elastomers have been introduced. In order to enable the CE and make composites more recyclable, through not just one but several lifetimes, the first challenge must be defining what is considered an acceptable definition of “recycling”. Are we including thermal recovery methods in this term? Are we considering energy recovery to be an acceptable end use? What do we mean by recycling: should we only consider upcycling, or is downcycling also acceptable? The answer to each of these questions, provides a slightly different perspective on acceptable disposal routes, especially for cross-linked based materials like thermosets and thermoset elastomers.

In considering mechanical reprocessing as higher on the disposal hierarchy than recovery, as it is now, then thermosets need to be chemically redesigned to “unzip”, to allow reprocessing. This technology is being actively researched and a recent award winning commercial use (discussed later) may help speed up developments. If, on the other hand, we value recovery by thermal methods, the recovered components from the original composite material need to be of equal economic value to maintain a value chain as long as possible. Growing the markets for these products and the products of mechanical recycling is an important component of developing CE systems. If we consider energy recovery to be an acceptable target, then we need to design composites with inherently higher calorific values and greater organic content. The same applies to thermoplastic systems.

Therefore, in considering any future strategy for composite materials within the confines of our current understanding and trends: a number of things should be considered.

Firstly, although out of the main scope for this article, the design phase plays a key part for the CE in ensuring future products are made that can inherently be reused, recycled and recovered through multiple process cycles. LCA will play a part in these initial design decisions. Currently we are still dealing with waste composites that were not designed with 2nd life in mind and will have legacy waste streams for at least another decade.

Standardization of both designs and materials to limit the vast material variations in the waste stream would be beneficial; however, the difficulties of applying this in such a highly customized field could be problematic. Big data and the creation of digital twin products (Qi and Tao, 2018) will help ensure there is traceability and confidence right through multiple service lives. Furthermore, there needs to be greater composite recycling infrastructure available, at logistically convenient positions and an expansion in markets for reuse, recycle and recovered fractions.

Other interesting developments for future composite recycling are:

- (1) Additive layer manufacturing (ALM).
- (2) Single polymer composites.
- (3) And perhaps most exciting – unzipping thermosets.

ALM presents new and exciting possibilities for future composite designs and distributed remanufacturing. Filled composite materials are now being routinely deposited, with an actively growing range of potential material choices. Whilst mechanical properties are still not anywhere at a level to compete with traditional manufacturing methods (Justo *et al.*, 2018), these

techniques are developing rapidly, becoming more common place in manufacturing factories, and will play a role in any future remanufacturing composite landscape, especially with the inherent digital components that come with this technology.

Single polymer composites result when both polymer and reinforcement are made up of the same material. These self-reinforced composite materials are commercially available, for example polypropylene based Curv® (www.curvonline.com, accessed 11/5/2020). In this case both polymer and reinforcement matrix are PP which negates the need for any separation process and allows for easier recycling.

Single polymer composites have been around for a long time and were first reported by Capiati and Porter (1975). With the CE in mind, these kinds of composites become increasingly attractive as reducing the materials count within a single material composite component to one enables much easier mechanical recycling. Work has been carried out in various other single polymer systems such as PET, Nylon 66, and polyethylene naphthalate (PEN) as well as PP and PE. These materials do not compete with the mechanical strength and durability properties gained from the use of conventional reinforcements such as GF, but they present a CE option wherever applications allow for lower inherent strength properties to be utilized. The reader is referred to a book by Mukhopadhyay and Adak (2018) for further coverage.

At the JEC Innovation Awards in 2020, Cobra International and Aditya Birla Chemicals' closed loop recycling of epoxy parts technology won an award. The products were water sport components (made by Starboard and Maui Fin Company), and products contained a novel recyclable epoxy resin (Recyclamine® technology) component. This allowed the use of a closed loop recovery process and the re-use of the recovered materials when the epoxy composite is recycled. Developments such as these using vitromers-thermoset technology, reviewed by Krishnakumara *et al.* (2020), allows for both recyclable and self-healing thermoset materials using triggers such as heat or pH to enable a break down in cross-links, rendering the thermosets back into thermoplastics. These can then be recycled by conventional mechanical recycling methods. This represents an exciting step-wise change in the approach to recycling of thermosets if sufficient control of environmental triggers to breakdown can be achieved right through to large structurally demanding products.

Conclusions

Moving forward, the idea of a CE has accelerated our ambition to improve composite recycling. Perhaps that is best illustrated by the development of vitrimer type technologies for thermoset resins. However, for a material such as a thermoset, the CE is still somewhat of a paradox for a science that has strived to create highly durable and enduring materials for highly demanding environments.

Further, what we consider as 'acceptable' in terms of the 2nd life product (downcycling or upcycling), cost and environmental impact are constantly developing. Economically viable products will always find companies to invest in them, but are they the most environmentally optimized solutions? How to we quantify the acceptability of the trade-offs?

Thermoset and thermoplastic composites and their elastomers, currently present two different waste scenarios. The problems facing thermoset waste, such as currently faced in the decommissioning of wind farms, aircraft structures and marine applications, still needs much further research and consideration.

Reuse provides some major challenges for thermosets, but also presents some intriguing possibilities. Can large thermoset structures find sufficient and local useful reuse applications in architecture and construction? Can they be turned into less demanding smaller components? Problems with structural integrity and consumer trust may need to be solved first. Further development in non destructive testing (not covered here) may hold the answers and allow these questions to be more fully addressed.

If we are able to unzip our thermosets at end of life, and they become thermoplastic in nature, this could be greatly beneficial to the CE. But can they be easily monitored and controlled, and again will consumers have trust? Considering the difficulties faced with controlled environmental degradation of polymers more widely, this technology will need to be proven effectively.

CF recovery methods are commercial examples of what can be achieved when economic viability, markets and environmental concerns align. However, there is still work to do in balancing recovery of both the polymeric component and the reinforcement (and fillers).

New markets for the recycled composites are still badly needed for ongoing commercial sustainability. Further, how will nanomaterial additives continue to impact on commercial markets and the environment, as our knowledge on their potential environmental effects grows.

There are still numerous unanswered questions for composite recycling to overcome, but the increasing use of big data could be used to give increased confidence in the provenance and traceability of composite components throughout entire cycles of life: through formulation, processing and onwards into service life and 2nd life applications, or 2nd process routes opening new markets and potential supply chains for products.

Although great progress has been made in the last decade, much work still remains. Finding the right blend of ecological, economical and technical solutions for composite materials is and will continue to be a great challenge moving forward this decade.

Sources of Further Information and Advice

"Recycling of composites and elastomers" covers a very wide field of research and therefore this article has only provided a brief overview of many very distinct and in-depth areas of research topics within this remit. However, the wide range of references should provide access points to further areas of interest.

Active commercial company websites have been included where appropriate, as well as useful links to wider groups of interest.

In terms of academic journals, it can be seen from the reference list that there are many that can be considered relevant to this field. Older references have been included where appropriate, to allow readers access to the original published information. It is recommended that individual papers and journals of interest are followed up that are relevant to individual sections within this article.

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Welcome to ELG.

Material Design-for-eXcellence Framework – Application to Composites

SPB Sousa and AJ Baptista, Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal

AT Marques, Faculty of Engineering of the University of Porto, Porto, Portugal and Institute of Science and Innovation in Mechanical and Industrial Engineering, Porto, Portugal

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Nomenclature

AI Artificial Intelligence
AR Augmented Reality
CFRP Carbon Fibre Reinforced Polymer
FRP Fibre Reinforced Polymer
GFRP Glass Fiber Reinforced Polymer

KPI Key performance indicator
LCA Life Cycle Analysis
M-DfX Material Design-for-eXcellence
MSM Multi-layer Stream Mapping
VARI Vacuum Assisted Resin Infusion
VR Virtual Reality

Glossary

Material Design-for-eXcellence (or M-DfX) New methodological approach to address simultaneously,

material properties and characteristics performance comparison via ratios, and eco-efficiency of material processes along the life cycle.

Introduction

Advanced materials play a key role in energy efficiency initiatives and policies addressing sustainability. In the case of composites, this role is globally recognised as instrumental to clean energy initiatives because of their structural performance and low weight that leads to a better fuel efficiency and, thus, greater environmental benefits fulfilling national and international sustainability, energy efficiency initiatives and policies. Due to these features, the pace of proliferation of these materials in manufacturing of complex aerospace, automotive, marine, sports, and energy structures are rapidly increasing ([Pervaiz et al., 2016](#); [Scelsi et al., 2011](#); [Tan et al., 2008](#)). In [Fig. 1](#) illustrates the ecosystem of the composites materials.

However, with the growth of sustainability challenges, the composite industries are seeking for holistic solutions, which ensure the reduction of the environmental impact, reduce time in the product development process, guarantees higher product quality, reduce overall costs and fulfil customers' requirements. To achieve all of these requirements, companies must use new models that ensure design efforts, customer, and societal needs are met from product ideation until its end-of-life. To do so, several companies are adopting Design for X (DFX) as an approach, which considers several requirements through different factors, Xs, to improve the product design as well as the design process ([Benabdellah et al., 2020](#)). The aim of this methodology is to make the product better suited for the different life phases and increase the general product performance.

Hence, the aim of this chapter is first to make a brief review of the state-of-knowledge regarding to design-for-excellence framework and how this methodology can be used in the composites industry. Second, a conceptual framework called, Material Design-for-eXcellence Framework – Application to Composites is introduced that addresses the design factors of each M-DfX dimensions and their associated modules.

State-of-Knowledge

Until now, very few studies in the literature have reported the concurrent-based design methodology for the development of composites products or processes.

State-of-the-Art “GAP”

Despite the availability of a number of material selection methods and tools, there is a lack of straightforward material performance methods that provide an easy and multi-dimensional assessment of the material properties relating to the inner structure of the material in a multi-scale proposition, supported by a Life-Cycle Assessment mindset ([Alemi-Ardakani et al., 2016](#); [Milani et al., 2011](#); [Rao and Davim, 2008](#)).

In this work, a novel framework entitled “Material Design-for-eXcellence” or “Material Design-for-X” – adopting “M-DfX” for abbreviation – is proposed. The M-DfX is an integrated approach and is proposed for a systemic assessment of a given material performance overall evaluation, where the Materials Properties are evaluated via dimensionless ratios with a benchmark mindset and in a multi-dimensional management form. Furthermore, the material performance is framed in conjunction with the manufacturing process and life cycle assessment, thus aiming to evaluate the overall material performance.

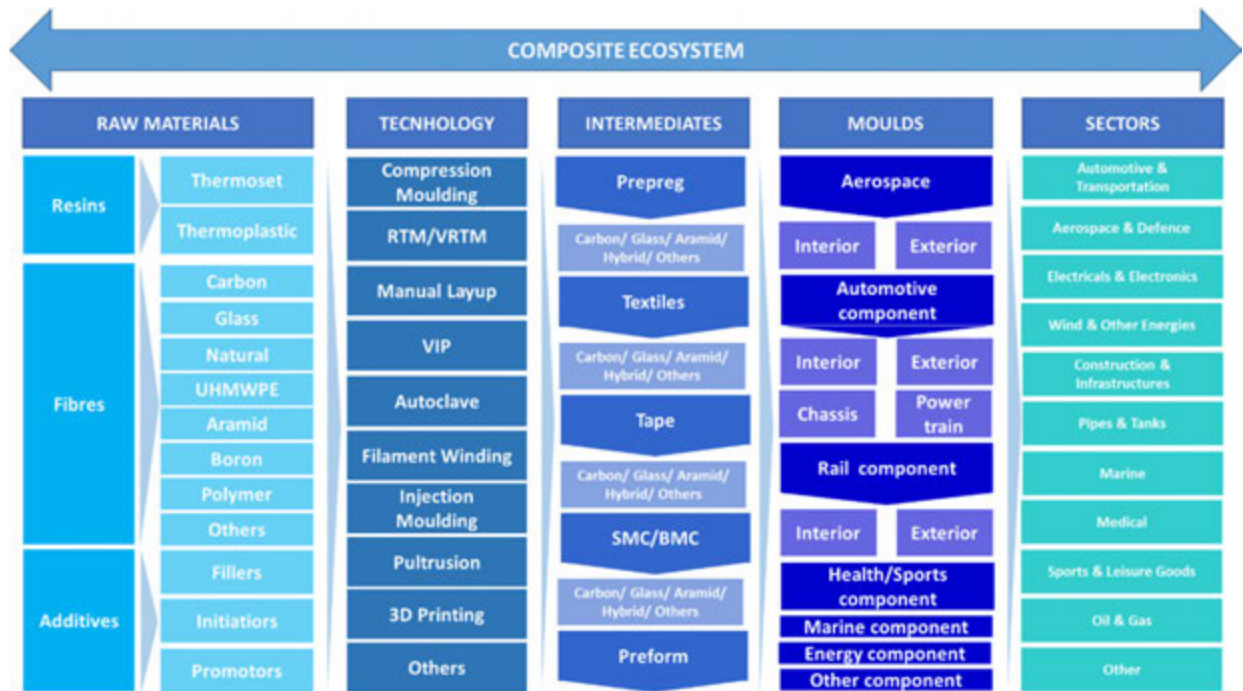


Fig. 1 Composites Ecosystem.

Material Design-for-eXcellence Conceptual Framework

The Material Design-for-eXcellence conceptual framework was created upon state-of-art methodologies developed by INEGI for complex product development multi-dimensional assessment (Atilano *et al.*, 2019) and production systems resource and operational efficiency assessment (Lourenço *et al.*, 2016). In the M-DfX framework, two fundamental aspects are assessed in an integrated way:

- (1) The material behavior and its multiple characteristics assessment, focusing on the effective measurement and benchmarking of materials. Here, the different types of characteristics are independently evaluated according to the material decomposition in different scales (e.g., macro-scale, mesoscale, and micro or nano-scale).
- (2) The manufacturing processes efficiency assessment (resources and also operations) – including the life cycle analysis (LCA) evaluation – to assess both resource efficiency involved in the manufacturing (either for raw-materials components and energy efficiency) and the environmental impacts.

By applying these two aspects, the material performance can be assessed in a holistic and life cycle-oriented form. On the one hand, the material properties' effectiveness is evaluated regarding thresholds or target values from benchmarking ("best in class approach"). On the other hand, the production efficiency (namely, for material and energy efficiency, thus the main resource usage) and LCA and perspective are also integrated to assess the cumulative environmental impacts.

The framework considers the analogy of product design holistic approaches – such as Design-for-X – to organize and assess the multi-dimensional performance for each "X" Material Property (Fig. 2).

The M-DfX framework was developed with the following main objectives:

- (1) Assess material performance in a multi-dimensional space for "X-Properties" in a systematic and more comprehensive way, comparing and benchmarking material properties in a normalized form;
- (2) Relate material properties performance with the material composition complexity in different scales (macro-, meso-, micro-, or nanoscale);
- (3) Integrate the material performance analysis with a Life Cycle approach, with key performance indicators such as Resource Efficiency, Environmental Impact Assessment, etc.;
- (4) Adopt a Lean Thinking approach, as for the use of visual management and waste identification in relation to production efficiency;
- (5) Cross-evaluation of material effectiveness assessment and material production efficiency via novel original scorecards and quadrant-like approaches; and
- (6) Support both material databases enhancement for a new perspective in material assessment and benchmarking, namely for material selection purposes, or support new material design and optimization.

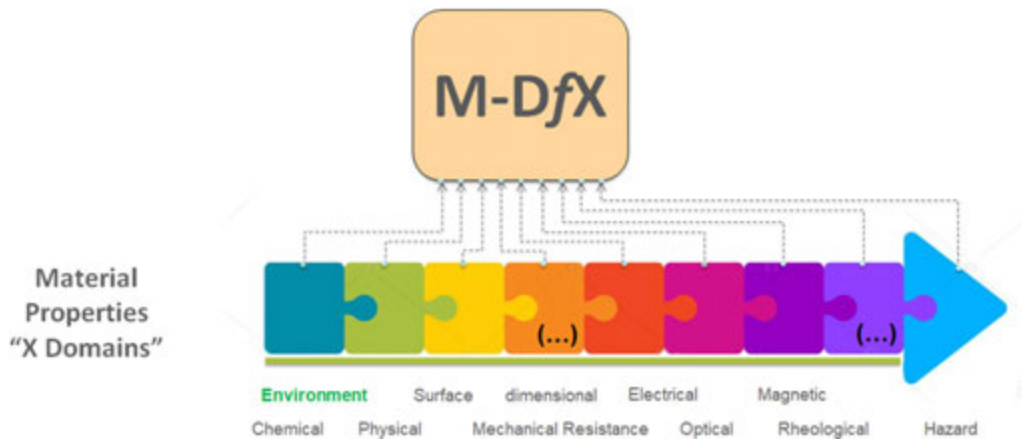


Fig. 2 M-DfX multi-dimensional overview regarding material life cycle and material properties “X” examples. Dimensional.

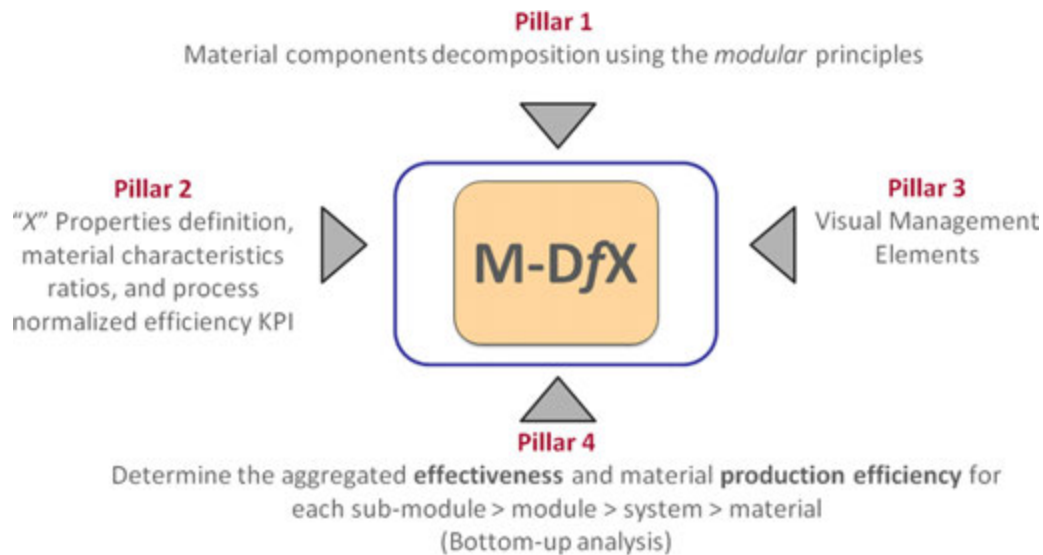


Fig. 3 M-DfX framework pillars.

Framework Description

The M-DfX framework is structured according to four fundamental pillars, which also correspond to the base sequence for the methodology formulation (Fig. 3).

The first pillar corresponds to the physical arrangement decomposition of the material. The objective is to apply the concept of modular design management and complexity management from the product-design environment into the material context. This can support a more understandable way to assess and detail the performance of the material and to facilitate the design stage of new materials. The M-DfX architecture allows the benchmark of multiple material properties (“X”) and characteristics. The level of material decomposition can be set in different scales – hierarchically related – according to the existing data and the purpose of the material characterization and application. The most common scales are identified: macroscale (e.g., for composite material structures), mesoscale, microscale, and nanoscale (Fig. 4). For indexation purposes, the tensorial form is considered for the “Material Component” decomposition into layers. For instance, for the three-layer decomposition, it assumes $M_{comp}(i,j,k)$.

The second pillar of the framework is the cornerstone to the approach since it holds a key part of the formulation behind the calculus for both the effectiveness aspect and efficiency and eco-efficiency assessment. For a given analysis – for instance, in a task of material selection, benchmarking, etc. – the user must first select the “X” properties that are relevant for a given analysis and, for each “X” property, the material characteristics (Mat_char) that are to be assessed and compared. The framework does not impose any specific classification of materials properties and associated characteristics, rather it recommends the most standard-wise approach from the state-of-art. For instance, regarding “X – Mechanical Properties” and “Mat_char – Creep rupture” the selected

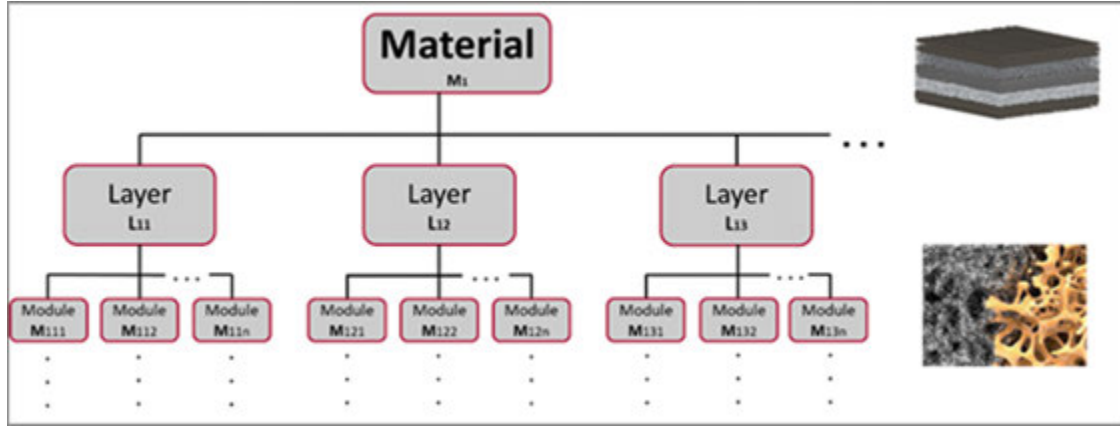


Fig. 4 M-DfX Pillar 1, material decomposition using modular principles.

characteristic indicator – to compare alternatives – is the effectiveness indicator, normalized according to the best creep rupture value for the materials selected in that study and a given material scale (e.g., macroscale or a material layer, etc.). Thus, for a given material component (M_{comp}), and for each material characteristic indicator ($Mat_{char,m}$) of a given X_n Property Domain, it is necessary to define the normalization target, either in relative form “Relative Best” (e.g., with the materials in a given current comparison) or absolute form “Absolute Best” (from the best-known material). For the normalization to “Relative Best,” the following equation can be used (Eq. (1)) for a given characteristic to maximize.

$$Mat_{char} \text{ RATIO} \{ Mat_{char}(X_{n,m}); M_{comp(i,j,k,...)} \} = \frac{Mat_{char} \text{ ACTUAL VALUE} \{ Mat_{char}(X_{n,m}); M_{comp(i,j,k,...)} \}}{Mat_{char} \text{ RELATIVE BEST} \{ Mat_{char}(X_{n,m}) \}} \quad (1)$$

For the normalization to “absolute best,” the same method is applied, substituting in Eq. (1) the reference figure of “Relative Best” to “Absolute Best” from a given Material Reference Database.

Due to the systematic normalization of the quantities of the M-DfX framework towards 100% – for cases where the material characteristic should be minimized – the equation terms should be inverted (Eq. (2)).

$$Mat_{char} \text{ RATIO} \{ Mat_{char}(X_{n,m}); M_{comp(i,j,k,...)} \} = \frac{Mat_{char} \text{ RELATIVE BEST} \{ Mat_{char}(X_{n,m}) \}}{Mat_{char} \text{ ACTUAL VALUE} \{ Mat_{char}(X_{n,m}); M_{comp(i,j,k,...)} \}} \quad (2)$$

For cases and material development activities where a given new material characteristic is better than the current assumed “Relative Best,” the new material replaces the former “Relative Best” so that the methodology remains consistent with the results scorecards (with effectiveness between 0% and 100%). An illustrative example of the M-DfX scorecard for effectiveness is presented in (Fig. 5), where the visual management attributes are represented for the four-color scheme and legend for effectiveness levels (in the example: red between 0% and 40%; orange between 40% and 69%; yellow between 70% and 89%; and green between 90% and 100%). The mathematical relationship of each M-DfX scorecard cell and the overall effectiveness is given below (Pillar 4).

To better compare the materials and support the decision-making for material selection, the M-DfX scorecards can be configured to filter higher aggregation levels for the analysis (e.g., Material level versus Property level in Fig. 6).

Besides the effectiveness assessment, the framework also maps and evaluates the efficiency, eco-efficiency, and resource efficiency of the material production, along with the macro-processes of the life cycle. To illustrate the assessment, it is considered the normalization (Mat_{EE_KPI} Ratio) of the (eco)efficiency KPI of each macro-phase in the material life-cycle (Mat_{EE_KPI} ; LC_Phase), considering the best reference values for the normalization targets (Eq. (3)). It can be “absolute best” according to the best materials in databases (as described above for the effectiveness formulas) – or the “relative best” if the normalization targets are filtered according to the set of materials in comparison to a given product application. Each KPI normalization can be applied according to each macro-phase so that a direct comparison of figures can be via done material vs macro-phase. Alternatively, the KPI normalization can be applied to the best figure of a given KPI of the life cycle of the material macro-phases.

$$Mat_{EE_KPI} \text{ RATIO} \{ Mat_{EE_KPI}(i); LC_Phase(j) \} = \frac{Mat_{EE_KPI_BEST} \{ Mat_{EE_KPI}(i); LC_Phase(j) \}}{Mat_{EE_KPI_ACTUAL \text{ VALUE} \{ Mat_{EE_KPI}(i); LC_Phase(j) \}} \quad (3)$$

In Fig. 7, the Material (eco)Efficiency scorecard is displayed for illustration, taking key macro-phases in material life cycle: raw-material extraction; process production; use-phase of the material incorporated in a given product/system; and EOL. These macro-phases can be adapted according to a given study with the level of detail in the analysis – as well as the selected KPI – but always in a representative way and consistently for all materials for comparison. In the scorecard, the column aggregation of the percentages is calculated as the average of the individual KPIs.

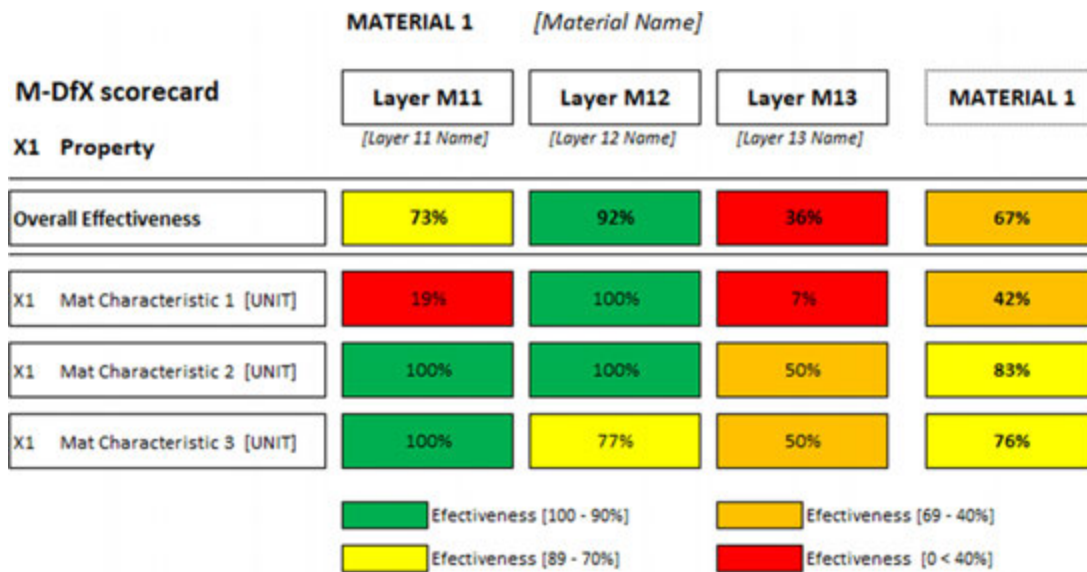


Fig. 5 M-DfX Effectiveness scorecard, illustrative example.

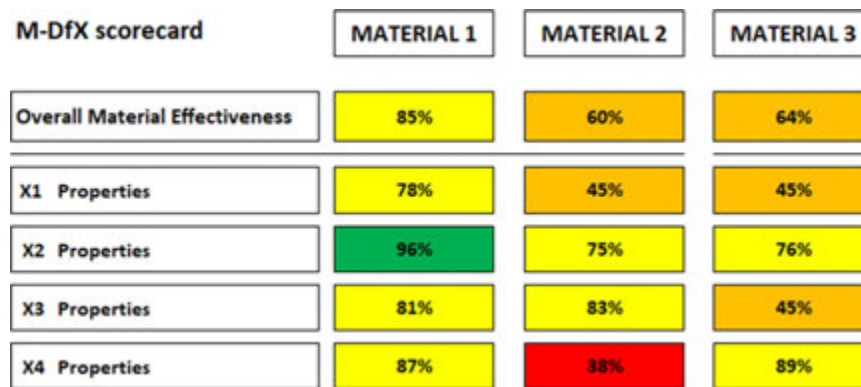


Fig. 6 M-DfX Effectiveness scorecard for comparison of different materials, for a given set of X Properties. Illustrative example.

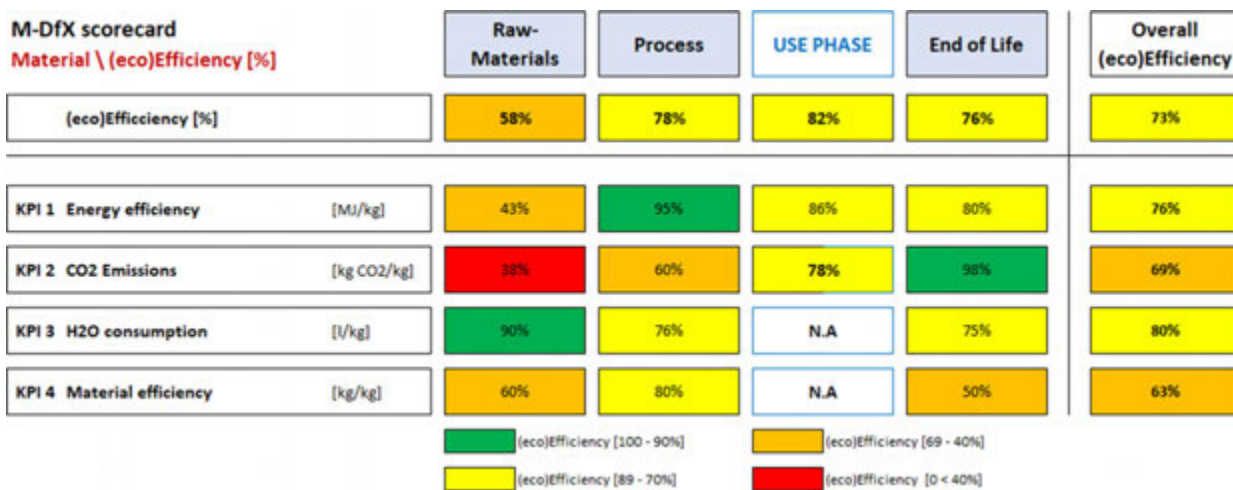


Fig. 7 M-DfX (eco)Efficiency scorecard for a set of KPI and macro-phases of the material life cycle. Illustrative example.

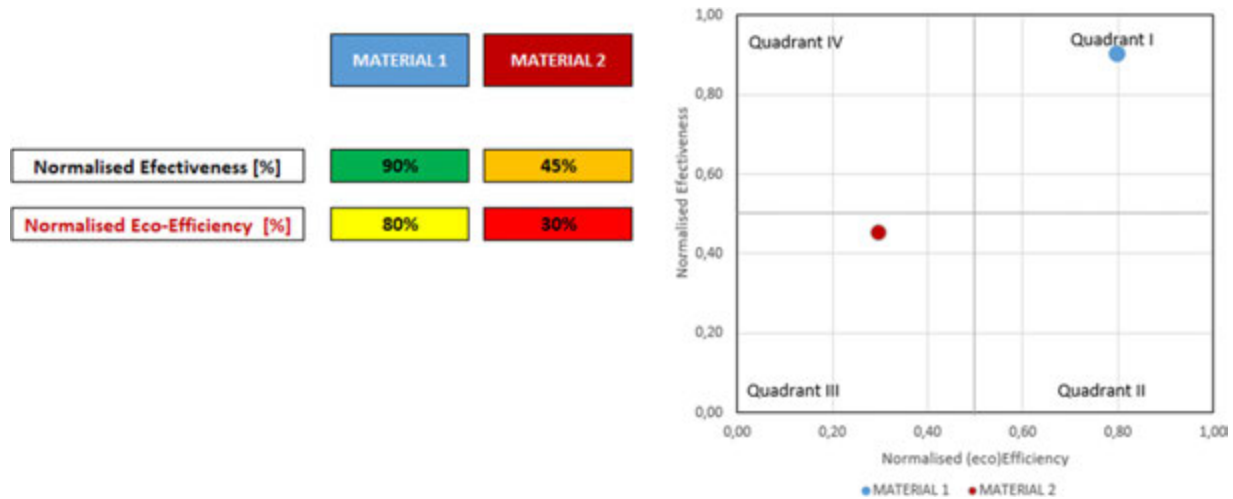


Fig. 8 M-DfX Quadrant Graphical results for overall performance materials comparison. Illustrative example.

Table 1 Main average properties of the main raw materials

Properties		Epoxy resin	Polyester resin	Glass fiber	Carbon fiber	GFRP	CFRP
Mechanical	Young's Modulus (GPa)	2.41	0.3005	78.5	242.5	21.5	109.5
	Tensile Strength (MPa)	67.3	15.35	1975	4600	189.5	800
	Elongation (% strain)	4.5	175	2.85	1.85	0.9	0.335
Thermal	Glass temperature (°C)	117	180	–	–	172	139.95
	Maximum service temperature (°C)	130	120	615	555	180	180
	Thermal conductivity (W/m°C)	0.1885	0.1575	1.275	140	0.475	1.94
Physical	Density (kg/m ³)	1255	1105	2575	1820	1860	1550
Cost	Price (€/kg)	8.79	3.485	2.095	25.15	25.15	33.85
Environmental (primary production)	Energy (MJ/kg)	128.5	71.35	51.8	286	–	–
	CO ₂ (kg/kg)	6.6	2.535	2.995	20.3	–	–
	Water (l/kg)	28	200	94.5	7.4	–	–

Note: CES_Edupack, 2019; Granta Design Limited.

Finally, the framework also considers a graphical comparison of the two performance components – effectiveness (assessing the properties characteristics) and (eco)efficiency (assessing the resources efficiency along the life cycle) – in the form of a Quadrant Graph for a more sensible overall comparison of the materials in comparison (Fig. 8).

Framework Applications for Materials and Manufacturing Processes of Composite Systems

The example proposed here is based on a vehicle that is used as an airport bus to transport people with their luggage between different gates and terminals. Most of these vehicles have aluminum bodywork; however, some manufacturers adopt fiber-reinforced polymers (FRP) (Machado, 2014). The production of the FRP is complex and involves several materials and production technologies when compared with aluminum. Table 1 presents the relevant properties of the main raw materials, based on the information of CES_EduPack (2019).

There are several processes for the Fiber Reinforced Polymer FRP production components, where the selection is driven by the design requirements of the application. One viable manufacturing process for FRP bus bodywork is vacuum-assisted resin infusion (Couto *et al.*, 2012; Khan and Mehmood, 2016). Table 2 lists the input data.

The use phase refers to the period of a vehicle component when it is functioning in its predetermined application. This phase dominates the energy consumption of vehicles, primarily due to impacts of vehicle weight (i.e., fuel economy). Therefore, the less weight of FRP is recognized in the transport sector in the use stage (Tapper *et al.*, 2020; Tempelman, 2011). Considering that the use of FRP could lead to a weight reduction of 10%–15%, for Glass Fiber Reinforced Polymer (GFRP) and Carbon Fiber Reinforced Polymer (CFRP), respectively, when compared to conventional aluminum bus bodywork, this could represent an energy consumption of 1.29–1.34 kWh/km (Couto *et al.*, 2012). Landfill has been the most common disposal route for composite materials as it is traditionally the most economical and it can easily handle large waste quantities. However, more recently, recycled processes have been developed

Table 2 Environmental impact of the process

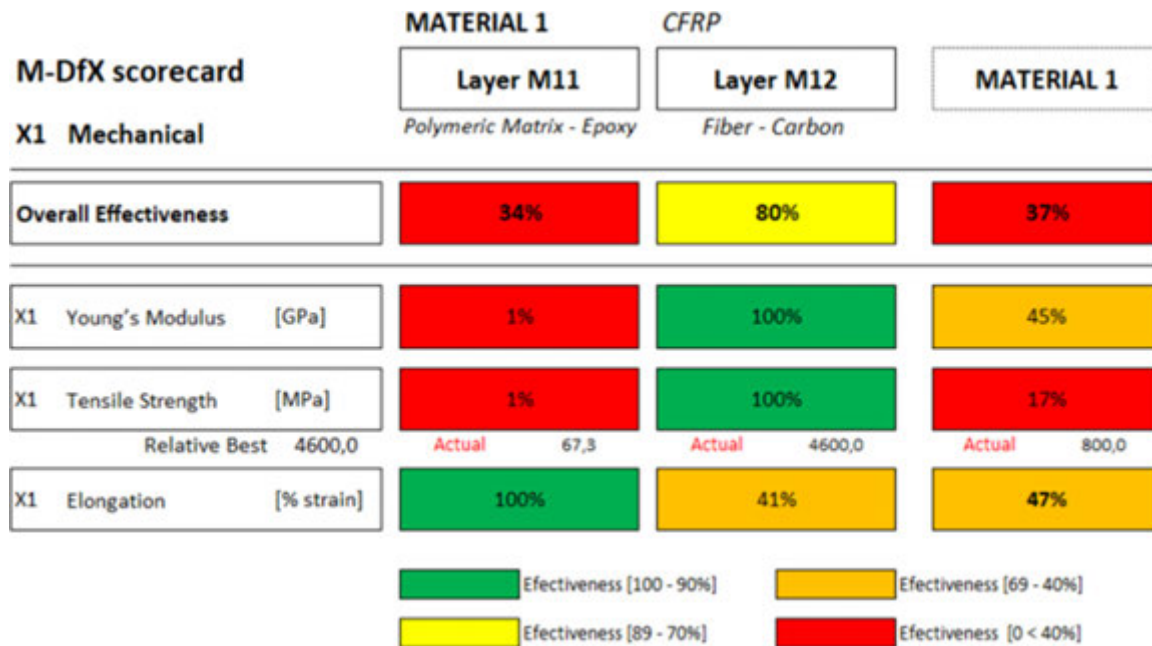
FRP	Vacuum-assisted resin infusion		
	Energy (MJ/kg)	CO ₂ (kg/kg)	Water (l/kg)
	10.205	0.817	11.18

Note: CES_Edupack, 2019. Granta Design Limited.

Table 3 Environmental impact of the main end-of-life treatments

Process	Energy (MJ/kg)	CO ₂ (Kg/kg)
Mechanical grinding	0.14–51	36
FB Pyrolysis	7.7–30	5.4–11
Solvolyis	15–64	–

Note: Tapper, R.J., Longana, M.L., Norton, A., Potter, K. D., Hamerton, I., 2020. An evaluation of life cycle assessment and its application to the closed-loop recycling of carbon fibre reinforced polymers. *Composites Part B: Engineering* 184, 107665. Available at: <https://doi.org/10.1016/j.compositesb.2019.107665>.

**Fig. 9** M-DfX Effectiveness scorecard for CFRP material, “X – Mechanical Properties”, for the two layers and aggregated result (Material 1).

and used for these materials, with different energy and CO₂ results (Table 3). The predominant treatments are mechanical, thermal, and chemical process (Li, Bai, and McKechnie, 2016; Rybicka *et al.*, 2016; Tapper *et al.*, 2020; Tempelman, 2011; Vo Dong *et al.*, 2015). Depending on the FRP recycling treatment type, it is possible to reuse either the fiber or the fiber and the matrix. The mechanical method is currently an industrial-scale process to recycle composite materials especially GFRP. Despite this practice, the environmental aspects of the technique – particularly its process energy demand – must be taken into account. In manufacturing, recycling and remanufacturing electrical energy demand dominates the environmental burden and global warming potential of this process (Gopalraj and Kärki, 2020; Grammatikos *et al.*, 2020; Shuaib and Mativenga, 2016). Pyrolysis and solvolysis are the most common process to recycle CFRP since these processes allow carbon fiber recovery with negligible surface damage (Gopalraj and Kärki, 2020; Kumar and Krishnan, 2020; Lopez-Uribeabarenechea *et al.*, 2020).

The “relative best” approach was adopted to normalize the effectiveness ratios for the (eco)efficiency along the macro-phases selected. Fig. 9 presents the results for CFRP material (Material 1) application to bus bodywork. When compared with GFRP for normalization, for the effectiveness in “X – Mechanical Properties” and the combination of the two layers (Epoxy polymeric matrix) and carbon fiber for reinforcement.

Regarding the Young's Module and Tensile Strength (T.S.) properties, a big difference in the ratio between the Carbon Fiber (Layer M12 of the material decomposition) and the Polymeric Matrix – Epoxy (Layer M11) is observed. The aggregation of the

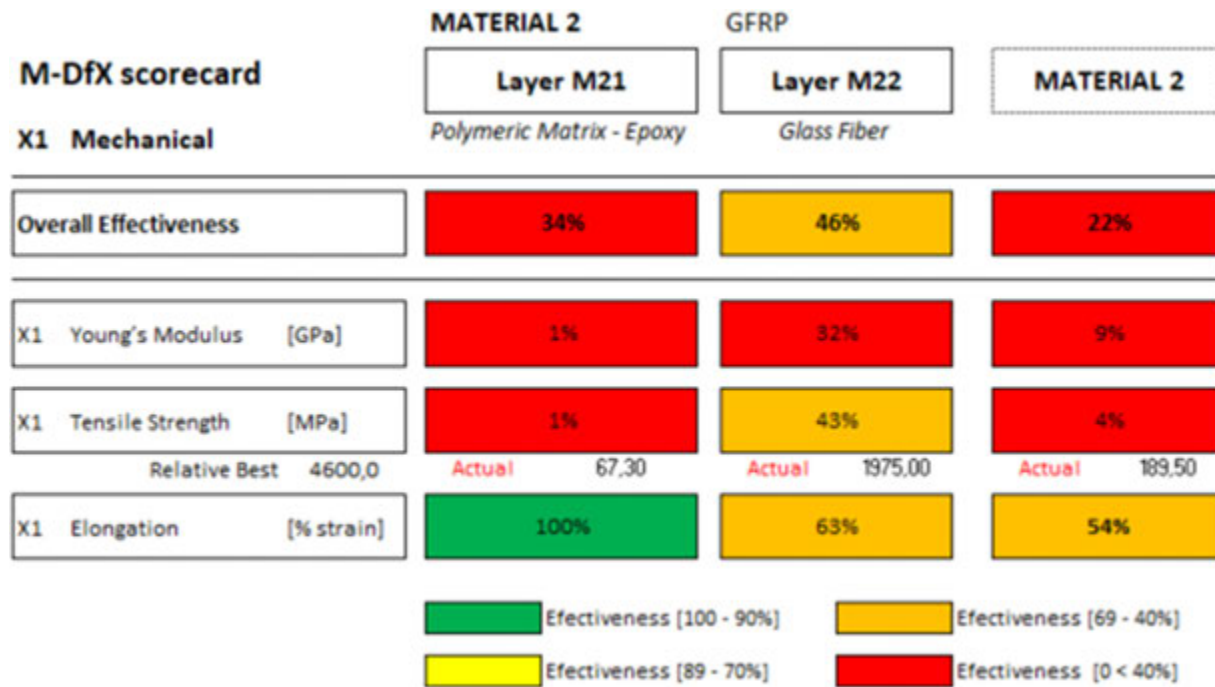


Fig. 10 M-DfX Effectiveness scorecard for GFRP material, “X – Mechanical Properties”, for the two layers and aggregated result (Material 2).

individual KPI results is made by line and it used the actual value of the composite material (e.g., 800 MPa for T. S.; by dividing by 4600 MPa from the Carbon Fiber, T.S. results in 17% ratio) for Material 1. The overall is computed by the average of the third column results (37%). For information purposes, the aggregated results for each layer (34% in Layer M11 and 80% in Layer M12) are displayed. For this case study, these three characteristics were selected since they were more relevant for the project design. For different sets of characteristics, the percentages will vary from those shown.

Fig. 10 presents the same configuration of the M-DfX scorecard as **Fig. 9**, but in the case of GFRP (Material 2) with targets for normalization relative to CFRP. For the Tensile Strength, the absolute values (MPa) are presented for better comprehension of the mathematical implementation of the above equations.

Besides “X – Mechanical Properties,” “X Thermal Properties” were also considered with the following material characteristics: maximum service temperature ($^{\circ}\text{C}$), thermal conductivity ($\text{W/m}^{\circ}\text{C}$), and glass temperature ($^{\circ}\text{C}$). Finally, a set of properties was considered: “X – Physical Properties” with material Density (kg/m^3) and “X – Cost” with material price ($\text{€}/\text{kg}$).

The aggregated results for all “X properties” and the two main materials (already integrated with the partial results of the layers) are presented in **Fig. 11** where the overall result is the direct average of the individual “Xi” properties for each material. The overall result is equal (40%); however, these equivalent 40% overall values are made up of different factors relating to mechanical and physical properties. CFRP is slightly better in Mechanical Properties and Physical Properties, but GFRP is better in Thermal Properties and Cost. The figures are generally low in percentage which means that there is a high deviation from the best reference of material components (between layers) and in absolute terms. For instance, cost percentage is affected by the lower cost of the polymeric matrix versus the cost of the fiber. No quantity weighted function was taken, but this could be studied further in the future to have more detailed results as a function also of the quantity or volume of material applied. One important asset of M-DfX is that it allows a detailed analysis of the material components and their impact on the integrated material to be applied in the product for each “X – Property” and characteristic (“drill-down” capability).

For the (eco)efficiency in this study intended as a set of eco-indicators for the macro-phases of the material processing and use across its life cycle, four main KPI were selected: energy efficiency (MJ/kg), CO_2 emissions ($\text{kg CO}_2/\text{kg}$), H_2O consumption (l/kg), and recyclability (%). Not all KPIs could be mapped for all macro-phases, either because of an absence of data available or because it is not applicable (N.A.). **Fig. 12** presents results for CFRP where the normalization was done phase by phase, according to the relative best reference material (CFRP and GFRP). The results are integrated column by column, assessing the macro-phase (eco) efficiency and the overall result is computed by averaging the macro-phase figures. The aggregation of the percentages could also be weighted as a function of the absolute value of each macro-process or other weight factor (cost or environmental impact). This will be explored in future works with the framework.

Fig. 13 shows the analog results for material GFRP. The CFRP performs slightly better than the GFRP, with significantly better results for the H_2O consumption in Raw-Materials extraction and CO_2 Emissions in EOL. Nonetheless, CFRP is clearly worse in Energy Efficiency in the Raw Materials phase. It was considered that both materials are processed by infusion and is the reason they share the same 100% result in the production phase. In synthesis, the M-DfX identifies that CFRP gets worse results in the

M-DfX scorecard	MATERIAL 1	MATERIAL 2
	CFRP	GFRP
Overall Material Effectiveness	40%	40%
X1 Mechanical Properties	37%	22%
X2 Thermal Properties	37%	43%
X3 Physical Properties	81%	67%
X4 Cost Properties	6%	8%

Fig. 11 M-DfX Effectiveness scorecard with integrated results for CFRP and GFRP material, for all X – Properties considered.

M-DfX scorecard	MATERIAL 1 CFRP				Overall (eco)Efficiency
	Raw-Materials	Process	USE PHASE	End of Life	
Material \ (eco)Efficiency [%]					
(eco)Efficiency [%]	60%	100%	100%	83%	86%
Energy efficiency [MJ/kg]	43%	100%	100%	100%	86%
CO2 Emissions [kg CO2/kg]	36%	100%	N.A	100%	79%
H2O consumption [l/kg]	100%	100%	N.A	N.A	100%
Recyclability [%]	N.A	N.A	N.A	50%	50%

(eco)Efficiency [100 - 90%]
 (eco)Efficiency [69 - 40%]
 (eco)Efficiency [89 - 70%]
 (eco)Efficiency [0 < 40%]

Fig. 12 M-DfX (eco)Efficiency scorecard for CFRP material.

Raw-Materials phase, but much better in EOL, compared to GFRP. In the Use-Phase, CFRP shows slightly better results, but GFRP also gives a very good result (96%). Like the effectiveness scorecard, M-DfX shows the trade-offs between the KPIs in analysis for each material. Fig. 14 shows the M-DfX (eco)efficiency resume scorecard.

Fig. 15 presents the Quadrant Graphical assessment, integrating both the effectiveness and (eco)efficiency for both materials from the overall M-DfX scorecards (Figs. 11 and 14). Despite the effectiveness result being the same for the two materials, for the selected “X-Properties” and characteristics, the (eco)efficiency of CFRP is slightly better. Overall, both materials fall into Quadrant II (good eco-efficiency m but poor effectiveness).

Prospective Developments

Identification of Future Process Needs and Opportunities

In the very near future, Material Design-for-eXcellence Framework must include social impacts, ethics, and an evaluation of social acceptance for all the stakeholders (material producers, product development, manufacturers, workers, logistics, and the end-users). In other words, we have to consider the view of Society 5.0, particularly if we want to pave the way for a super-smart and environmental-friendly, healthy, and safe society. This means that artificial intelligence (AI), cyber-physical systems, virtual reality, augmented reality, and data analytics will go beyond current approaches (Rauch, 2020; Roblek et al., 2020).

Bearing in mind the need for a sustainable planet, a new paradigm for product development will have to appear. It will be required that the products have a longer life and are able to receive new technological updates. Hence, the M-DfX Framework will have to include a design for update. The efficiency and efficacy of the products must consider the possibility to replace and add modules to an old product. It will have also to consider logistics – in other words, minimize product “travel” to reach the consumer.

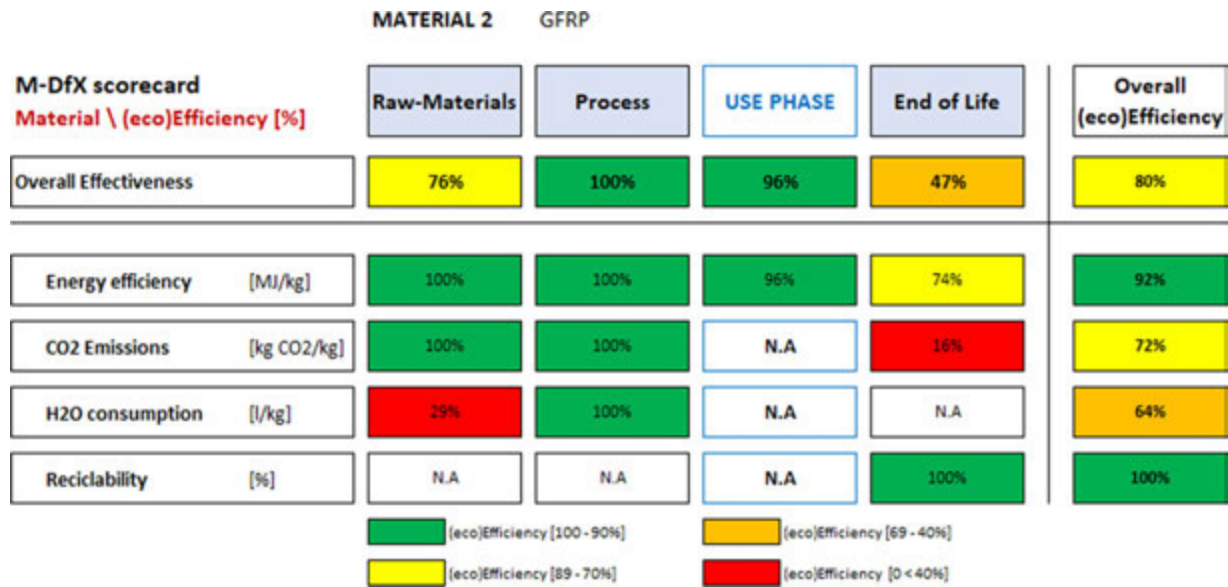


Fig. 13 M-DfX (eco)Efficiency scorecard for GFRP material.

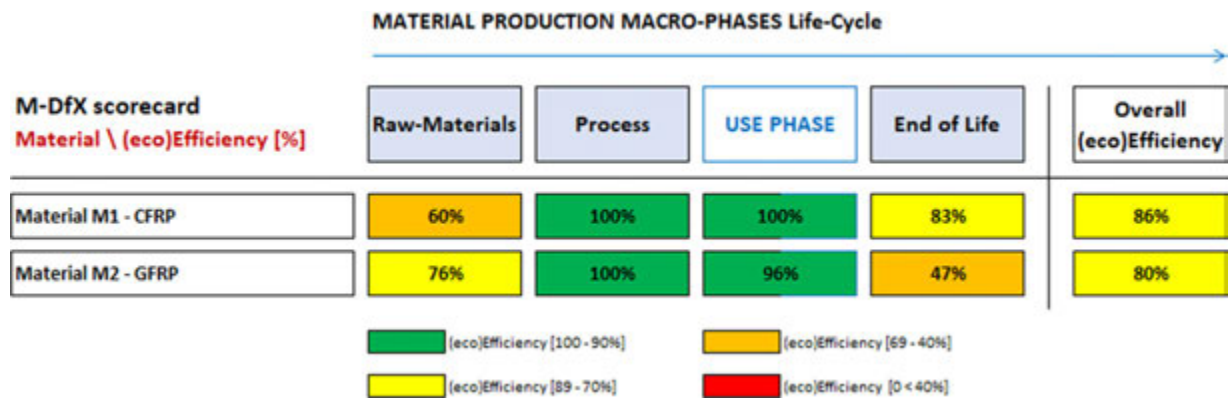


Fig. 14 M-DfX (eco)Efficiency scorecard resume, for both materials (CFRP and GFRP).

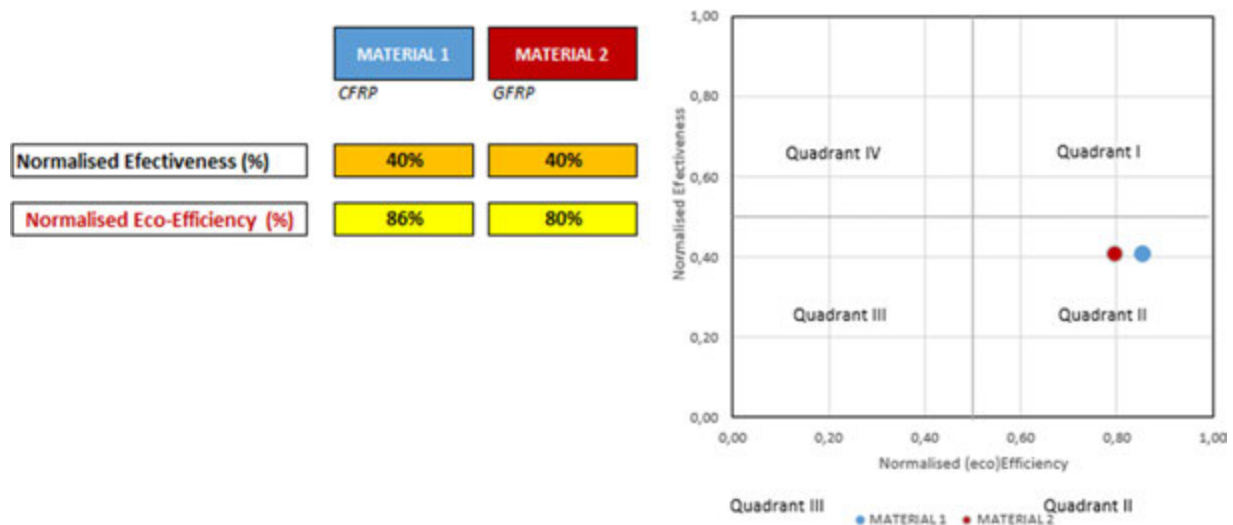


Fig. 15 M-DfX Quadrant Graphical assessment, for both materials (CFRP and GFRP).

Markets will be more fragmented with the reduction of “extractivist”. The world needs to look for solutions that use less and less materials extracted from non-renewable nature. The circularity of products and materials must be present in this framework.

A high potential manufacturing process (i.e., additive manufacturing hybridized with subtractive technologies) and an innovative product – high-performance composites parts produced without molds and with tailored properties – present a new challenge. Both processes and products hold a high potential of possibly converting tradable goods to foster industrial growth. 4D printing of composites (i.e., achieving a shape upon curing of the composite system) gives the possibility to create products without molds, which will introduce a challenge in M-DfX Framework.

M-DfX will have to deal with the development of expeditious ways of creating hybrid profiles and sheets of metal alloys, preferably aluminum alloys, magnesium alloys, titanium alloys, steel, or combinations thereof, and a fiber-reinforced polymer matrix.

Cyber-Physical Systems will have to be implemented to create new business models involving the following:

- Customization
- Communication Technologies
- Internet of Things
- Process simulation
- Remote sensing, self-sensing, and morphing
- Particle swarm optimization
- Genetic algorithms
- Fuzzy logic controller
- Artificial Bee Colonies.

5G automation, sensors, data, and AI are tools to be further exploited, not an end.

Further Studies of the Material Decomposition (Macro-Meso-Micro-Nano)

Further studies on multi-physics, multi-scale simulation, and mathematical models as a bridge between the physical and cybernetic worlds – in the form of Material Digital Twin – will allow the framework M-DfX to be explored in its full potential. Digital and digital twin will develop the best-fit constitutive laws (Glaessgen and Stargel, 2012; Tao *et al.*, 2018).

Digital Models will assist in the actual implementation of:

- Deep learning
- Twin-based prediction of best tool correction strategies
- Prediction of sensitivities
- Experiences with self-learning evaluation methods
- Best selection of hardware components – Control strategies for smart processes
- Self-learning quality control
- Optical detection of surface defects.

Bearing in mind that researchers are creating more and more new materials that are small (picomaterials will make nanomaterials look like giants), fast, and have diverse properties at different scales, Material Design-for-eXcellence must be considered at increasingly smaller scales.

Artificial Intelligence Application for Material Design Optimization

AI, with deep learning algorithms, will enhance the possibilities of creating metamaterials, explore functionality, new geometries, and adjustable properties. Hence, it will be possible – through a computational, data-driven approach – to invert the design process and develop a new selection of materials with targeted properties at different scales (Caldas and Norford, 2002; El Majdoubi *et al.*, 2020).

Theory-guided machine learning, being an emergent approach integrating domain knowledge and machine learning, can simulate aspects of the full composites processing chain, numerically giving efficient tools (Liu *et al.*, 2020; Wagner and Rondinelli, 2016). When M-DfX became a software module, the above technics can be included in evaluating designs for manufacturability related to materials, with speed and accuracy.

Conclusions

Material Design-for-eXcellence Framework presents an original approach to address the challenge of evaluating material performance with a multi-dimensional assessment for the material properties, relating to the inner structure of the material in a multi-scale proposition and supported by a Life-Cycle Assessment mindset for different macro-phases (Raw Materials extraction, Pre-processing, Processing, Use-Phase, and EOL). M-DfX formulation encompasses a systematic and visual way – through the evaluation of material properties (“X” dimensions) and related characteristics in a normalized form. It manages the material composition complexity in

different scales, adopting a modular configuration analogy. The integrated analysis of material performance is attained via an effectiveness assessment of the properties' characteristics cross-evaluated with efficiency/eco-efficiency aspects, within a Life Cycle approach, resulting in new original M-DfX Scorecards and Quadrants Graphs tools. It has adopted Lean Principles – such as for the use of visual management and waste identification in relation to production and resource efficiency. A demonstration example of M-DfX was given for the framework demonstration in the Composite Materials field, comparing a CRFP composite versus GRFP composite real use case application for the body of an airport bus vehicle. Future developments and research lines for M-DfX were also described, for instance, by its use for New Material Design and optimization by means of AI algorithms.

Acknowledgments

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Joining of Composite Materials: An Introduction

Antonello Astarita, Department of Chemical, Materials and Industrial Production Engineering, University of Naples Federico II, Naples, Italy

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The joining of composite materials is a topic of primary interest for industrial applications of these materials, what is more is a complex topic that requires multiple expertise to be fully understood. In the last years the joining of composites with metals, ceramics or concrete is also gaining an increasing interest thanks to the development of tailored hybrid structures. This section aims to provide an overview of the technologies to be used for the joining of composites, also including a summary of the facilities required and of the performances that can be achieved.

The first article co-authored by Antonello Astarita, Roberta Della Gatta and Alessia Serena Perna entitled: "Introduction to the joining of composites" introduces the problem of the joining of composites. Firstly, the concept of the joining itself has been defined then the various technologies to date adopted for the joining of the different typologies of composites have been introduced and presented. Particular attention has been paid to define a categorization of the joining techniques based on the different typologies of composite materials. This article aims to be a kind of atlas of the different joining techniques and a map for both practitioners and researchers who need to approach this field.

The article co-authored by Ricardo Carbas, Eduardo Marques, Catarina Borges and Lucas da Silva entitled: "Joining of polymer matrix composites (PMCs) – adhesive bonding" offers an overview on the adhesive bonding of polymer matrix composites. They studied the most used techniques to produce these joints and then discussed the main applications. In the article the mechanical behavior of the joints is also presented and discussed. The authors investigated also dissimilar joints, for instance the polymer to metal joining that can be extremely useful for some applications. The article provided also some design guidelines and some indications for future research.

The article authored by Mariana D. Banea entitled: "Dissimilar joining of PMCs to metals – adhesive bonding" tackles in a very detailed manner the problem of joining PMCs to metals. This article gives an overview of recent advances in adhesive bonding joining of PMCs to metals and highlights the main issues and challenges in joining dissimilar materials. The main parameters affecting the strength of dissimilar adhesive joints are discussed. Further, several methods that have been used to predict failure in dissimilar bonded joints are briefly described. The influence of the environment on the in service joints has been also investigated and some guidelines have been released.

The article co-authored by Pietro Russo, Ilaria Papa and Valentina Lopresto entitled: "Mechanical properties and non-destructive evaluations of joints based on polymer composites" presents an overview on the mechanical properties of joints. In particular, the influence of chemical-physical, geometric and environmental factors that mainly affect the efficiency of the bonded joints and, therefore, influence their characteristic failure modes and durability is discussed. Moreover, an overview of the main mechanical and non-destructive characterization techniques usually employed to validate the performances and/or to monitor the integrity over time of joints based on FRP composites is reported.

The article co-authored by Mahesh V.P., Sooraj Patel, Anurag Gumaste and Amit Arora entitled: "Joining of polymer matrix composites through friction stir processes" describes in details the joining of polymer based composites through friction stir welding. The physical basis of the process are described in details, the effect of main process parameters on the properties of the joints are also discussed. The influence of the tool design is also presented. Within the article is also discussed the modeling of the joining process and the needed fixtures are also presented. The equipment required are also shown and a literature survey, reported in the article, presents the latest results achieved in this field.

The article co-authored by Justo J., Barroso A., Blazquez A. and Paris F. entitled: "Joining of PMC to concrete for structural applications" describes the processes used to achieve a structural bonding between PMCs and concrete. The article describes all the aspects of these joints: the surface preparation of the parts to be joined is carefully studied then the different joining techniques are presented. The article also includes a section dealing with a discussion of the properties of these joints. Moreover numerical simulations are introduced to present and describe the in service behavior of the joints.

The article co-authored by Antonio J. Gamez, Severo Fernandez-Vidal, Alvaro Gomez-Parra, Pedro Mayuet and Ana Valerga entitled: "Mechanical Joining of Stacks" is devoted to stacks, hybrid structures built by assembling layers of material to enhance their characteristics. To begin with, a formal classification of hybrid structures according to the material is developed. Then, the main three different methods of joining: mechanical, chemical and thermal, are explained with their advantages and drawbacks, focusing on the mechanical joining of stacks. Finally, some of the main sectors where we can find this type of structures are commented.

The article co-authored by Juan Manuel Vazquez-Martinez, Irene Del Sol, Jorge Salguero, Moises Batista and Carlos Ramirez Alcala entitled: "Mechanical joining of composites: drilling related aspects" is devoted to the study of the drilling of composites to prepare the parts for mechanical joining. The article covers all the aspects of drilling, from the drilling techniques to the properties of the drilled parts. The influence of the drilling on the mechanical properties of the joints is also discussed and explained. Some guidelines are also released and explained.

Introduction to the Joining of Composites

Antonello Astarita, Roberta Della Gatta, and Alessia Serena Perna, Department of Chemical, Materials and Industrial Production Engineering, University of Naples Federico II, Naples, Italy

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Introduction to Joining

A joint is defined as a material or a device capable of connecting distinct structures. If the joint is properly realized, the loads can be transferred from one side to the other, acting as a single structure and possessing suitable rigidity for the proposed application. The advantages of using assemblies fall within the chance of overcoming some limits related to the choice of a single-piece structure: high functionality and manufacturability and reduction of costs. Nevertheless, the transmission of loads between structural elements is a key problem in the design of the structures and is especially crucial for composite materials (Matthews, 1987). As known, the joining of structures implies the joining of different materials and, for inhomogeneous materials like composites, this issue can be particularly challenging (Worrall *et al.*, 2020). The fact that composites are often manufactured as a single part could diminish the implementation of joining. However, there are different valid reasons to approach this process. For composite materials, joining can (1) limit the utilization of materials, (2) overcome the size and shape limitation related to the fabrication process (3) allow disassembly for repairing and maintenance operations (4) facilitate responsible disposal (5) allow joining of diverse kinds of materials which cannot be joined with conventional methods. The methods applied for joining composite materials are like those applied for joining isotropic materials. However, the strong directionality of their mechanical properties results in low strength through the thickness of the bonded structures, causing peculiar issues that have to be carefully considered when designing load-carrying joints (Tierney *et al.*, 2000). Often, the full strength and stiffness characteristics of the laminate cannot be transferred through the joint without a significant penalty. Thus, the topic of fastening devices is critical to the successful use of composite materials. This section will focus on different joining methods for composite materials and their implementation for several types of composite.

Different Joining Methods

Depending on the main forces employed to form the bonding between the structures, the joints can be allocated into three main categories: mechanical (better known as fastening), physical (usually referred to as welding), and chemical joints (adhesive bonding) (Messler, 2004) Fig. 1.

Mechanical Joints

Mechanical joining is a process that employs the use of a device (called a mechanical fastener) to mechanically join (or fasten) two or more objects together. Mechanical joints use compressive residual stresses throughout the joint to keep the components in contact and necessitate the balance of the tensile stress away in the system (Messler, 2004). Fasteners are widely used in structures with high load-carrying capability due to their reparability and replaceability. The most used fasteners are screws, rivets, and bolts (Jones, 1998), as portrayed in Fig. 2. Screws are manufactured with a head on one edge and threads on the other. Bolts are like screws except for the fact that they involve a nut at the side of the thread. A rivet is a short metal pin or bolt with a headless end being beaten out or pressed down when placed and its installation is irreversible. The variable head shapes can be flat, buttoned, semi-spherical, or other geometries. Rivets are mounted with special tools that plastically deform the ends to ensure mechanical fixing (Thomas, 1982).

Mechanical fastening is particularly suitable for the bonding of different materials or creating temporary repairs and it does not require particular surface preparation (Troughton, 2009). However, an increment in terms of weight is an inevitable drawback due to the use of fasteners.

The chosen materials to produce those fasteners highly depend on the fastener/laminate compatibility to avoid coupling issues such as galvanic corrosion. The most widespread choice is a metallic material, but in the last few years, many studies focused on the manufacturing of CFRP fasteners (Mosallam *et al.*, 1994; Yun *et al.*, 2008). Even Though there are various advantages related to the mechanical joints, some difficulties related to the presence of the hole can be evidenced. When using the mechanical fastening, the realization of holes in the samples is required before the joining process, and, in the case of composite the fibers can be damaged and that causes an increase in stress in the hole (Hallett *et al.*, 2009; Bhattacharyya and Horrigan, 1998). It is widely known that the creation of holes in a directionally reinforced material weakens the composites. Also, drilling the holes requires controls to avoid the delamination of the laminate and consequently separation of the plies and to avoid damages of the tool caused by the high abrasive reinforcement (Brinksmeier *et al.*, 2011). Due to the characteristics of the composites, the load transmitted via fasteners can lead to a stress concentration 6 ± 10 times higher than isotropic materials around the hole and may lead to the premature failure of structures. In fact, different from ductile materials, composite laminates are non-homogenous and anisotropic, and mostly brittle (Hoffman, 1967). Consequently, the main drawback associated with the practical implementation

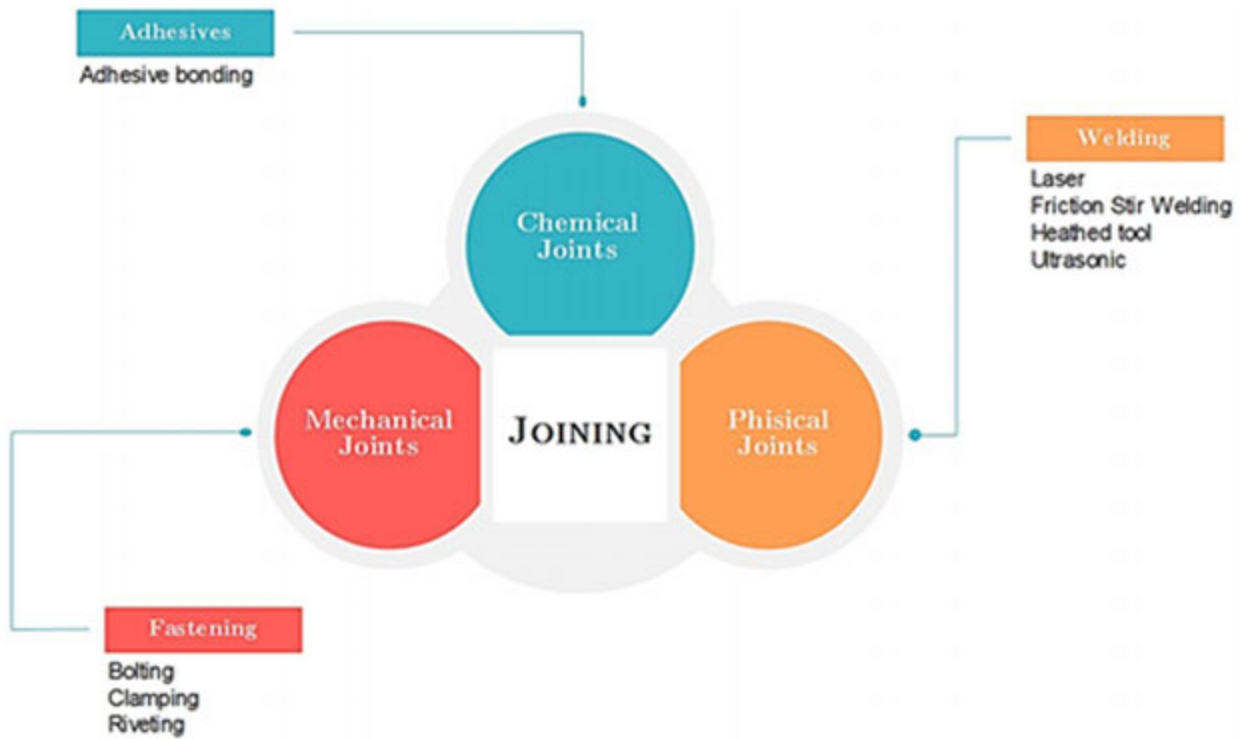


Fig. 1 Main typologies of joining techniques for composite materials.

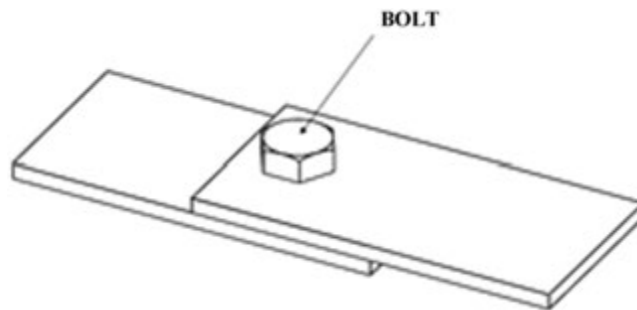


Fig. 2 Example of a bolted joint.

of composite bonded joints is the high possibility of failures, especially with delamination, of the substrates, which does not depend on the joint material. Also, with thin structures, the tear-out or pull-through of the fasteners can be observed.

Physical Joints

Physical joining techniques are processes that allow joining materials by transferring energy to the parts to bond or a third material (i.e., brazing or soldering) and they are usually known as welding techniques (Connor *et al.*, 1987). In the physical joining process, parts are made to join along their contacting surfaces through the application of heat or pressure, or both. The energy may cause the materials to melt (as in the most common welding techniques) or to soften without reaching the melting temperature (as in the friction stir welding technique). According to the heat input, these processes can be grouped into:

- (1) *Fusion welding.* The materials are heated up above the melting temperatures to be welded together. In this case, the two involved materials or a third material is melted to create the joint. According to the heat source, the fusion welding process can be divided into three main categories: arc welding, gas welding, and high energy welding processes. The most used technique to join composites are:
 - *Laser-assisted welding technique.* This technique, suitable for joining both sheet film and molded thermoplastics, a laser beam to melt the plastic in the joint region. The laser generates an intense beam of radiation (usually in the infrared area of the electromagnetic spectrum) which is focused onto the material to be joined (Katayama, 2013).

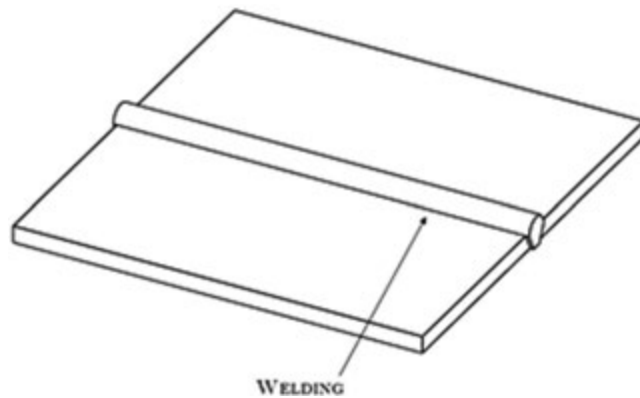


Fig. 3 Example of welded joint.

- *Hot plate welding.* The parts to be welded are held in fixtures, which press them against either side of a heated plate. Once the parts are sufficiently molten, the platen is removed. The components are then pressed together and held until they are cooled (Grewell and Benatar, 2007).
 - *Brazing.* In the case of brazing, two or more pieces are joined together by melting and flowing a filler metal into the joint section, the filler metal having a lower melting point than the adjoining metal (American Welding Society, 2007).
- (2) *Solid-state welding.* The process occurs below the melting temperature of the components; the contact flaps are subjected to high pressure and are heated up at the same time. These techniques rely on different multi-physics phenomena, and consequently, they can be divided into three main groups:
- *Diffusion bonding.* This is a solid-state process where an atomic diffusion takes place between two similar or dissimilar materials (Kazarov, 1985) usually conducted in a vacuum to keep surfaces clean and oxide-free. This technique is mostly used for welding mini or microchannel devices, commonly employed for biomedical implants, nozzles, and other precision assemblies, and for devices for high-temperature application where the softening of the material can cause the joint's damage.
 - *Friction welding.* In this case, the welding is achieved by the heat of friction between two surfaces. The most used friction welding technique is Friction Stir Welding (FSW) (Mishra and Ma, 2005). With this technique, a rotating tool is used to generate heat for welding. The advantages of this joining technology are that it is automatable, cheap to assemble and use, and does not need any extra added material during joining. Disadvantages are that the tool leaves a mark along the path it runs through and that the metal specimen is under significant mechanical stress during the joining process.
 - *Ultrasonic welding.* This technique involves the use of high-frequency mechanical sound energy to soften or melt the thermoplastic at the joint line (Tomiyasu and Takahashi, 1963).

In the case of physical joining, the interface zone is physically and chemically distinguishable from the joining parts and is called a welding bead. The usual welding configuration is shown in Fig. 3.

The key feature of those techniques is that the transfer of energy between the parts causes the materials to mix at the interface guaranteeing a strong bond (Ramakrishnan, 1972). It appears evident that those techniques are more suitable for materials able to soften or melt when energy is supplied. For this reason, thermoset-based composites are not suitable for those techniques as they cannot be melted and reshaped (Todd, 1990). However thermoset composites can be joined by melting a thermoplastic interlayer to create a joint between the parts, but, as there is no intermixing of the materials at the interface and no molecular diffusion, this technique can be ascribed as adhesive bonding rather than a form of welding (Messler, 2004). It is important to notice that usually for every typology of composite materials, the matrix and the fibers do not reach the melting or softening point at the same temperature. For this reason, the matrix materials rule the characteristics and suitable parameters for the joining process, while the fibers remain in a solid-state. The usual defects observed when welding unreinforced materials may still be present in the welding of composite materials. However, an ulterior plethora of issues may occur due to the presence of the reinforcement (such as segregation phenomena of abnormal interaction between matrix and reinforcement).

Chemical Joints

Adhesives are the most versatile among all the presented joining methods. Adhesives can be suitable for different types of materials, including metals, ceramic, and a combination of them, and they are commonly employed in aviation, automotive, and building industries (Schwartz, 2010; Higgins, 2000). The process of the chemical joining by using the adhesive is called adhesion bonding. To join two materials, referred to as adherents, a layer of adhesive is inserted in the region of interest and let cure. In some cases, a primer is applied to the surface to improve surface properties and to ensure a more effective connection (Adams, 2005). Before bonding, cleaning of the adherents' surfaces is required to remove grease, impurities, or even oxides, usually with mechanical polishing (Ebnesajjad, 2013). Depending on the workpiece and on the joint application, one or more adhesives can be used. The properties of the adhesives are chosen according to the specific application; normally epoxy resin but also acrylic,

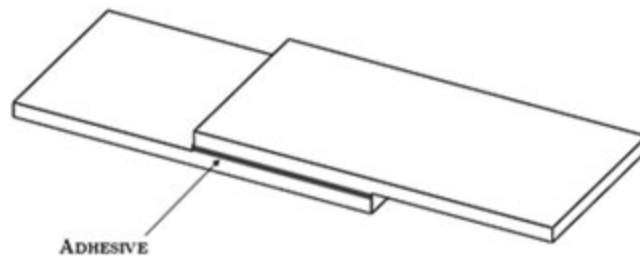


Fig. 4 Example of chemical bonded joint.

phenolic, or polyurethane are used and presented in liquid, paste, or film form (Adderley, 1988). Of course, the joint strength is related to the joining surface area. This technique can be considered a surface-driven process where stress and strains are transferred across the adhesive interface between the two laminate planes, as schematized in Fig. 4.

The adhesion bonding provides several benefits over conventional joining methods, such as continuity of the distribution of loads and stresses within the structures and lower weight and cost (Higgins, 2000). Compared to mechanical fastening, there are fewer risks for the joining of thin structures and no galvanic corrosion can occur between the dissimilar parts.

The main issues are the complex preparation of the surface and the geometry of the joint, the combination of the materials parameters of the adhesive and the adherents, and lack of strength and durability in different environments (i.e., higher temperatures) (Watts, 2010). Also, it is difficult to disassemble the ensemble and the repairing of the joint.

Joining of Composites

As introduced in the previous paragraphs, depending on the materials to join, one of the three categories of processes could be more suitable for the application. Thermo-sensitive materials like polymer-based composite are not suitable for techniques involving elevated temperatures (such as fusion welding or hot plate welding) while chemical bonding results in low-bond strength when applied to ceramic-matrix materials. For this reason, it is necessary to deeply analyze the most used joining techniques for different typologies of composites. In Table 1, the most suitable techniques used for the various typologies of composite materials are portrayed in approximate order of frequency of use.

Joining Polymer Matrix Composites

Polymer matrix composites, known as PMCs and FRPs, are the most commonly used composite in the industries (Abramovich, 2017). Depending on the polymer matrix type, FRPs can be divided into two types: thermoplastic and thermosetting matrix based. The most common technique for joining PMCs is adhesive joining (Tierney *et al.*, 2000). For PMCs, structural adhesive joints provide several benefits over conventional joining methods, such as more uniform stress distributions, higher resistance to fatigue, and the possibility of joining dissimilar materials, which can lead to structures with reduced weight and cost. Besides, high-performance structural adhesives are a key factor behind the growing industrial use of composite materials (Dillard, 2010). Usually, the adhesive used is similar to the matrix of the composite thermosetting polymer-based adhesives are generally used for the bonding of thermosetting-matrix composites while thermoplastic polymer-based adhesives are generally used with thermoplastic-matrix composites (Messler, 2004). One of the problems encountered in adhesive-bonded joints is joint distortion due to bending of thin adherents under eccentric loading and/or the inability of the adhesive to comply elastically or plastically and that requires different geometry of the adhesive surface.

As for mechanical joining techniques, as mentioned above, one of the main issues is the weakening of the composite through the drilling of the hole (Brinksmeier *et al.*, 2011). Also, certain reinforcements can be highly abrasive to cutting tools, including drills. The implementation of screws is limited because they are susceptible to pull-out and thread stripping especially from softer matrix materials, such as thermoplastic, while bolt and rivets assure more stability of the joint. Anyway, no substantial change in the micro- or macrostructure of the polymer material occurs during or after the joining process. The presence of external parts also increases the weight of the joint.

Regarding physical techniques, they are mostly applied for thermoplastic-based composites (Todd, 1990). The welding process involves the melting of the matrix polymer alone and not of the reinforcement. The main advantage of welding is the high flexibility of the process. Different from mechanical fastening, the use of welding reduces stress concentrations in the holes and additional joining elements are not necessary. The fusion of thermoplastic composites offers many advantages over mechanical fastening and adhesive bonding because thermoplastics can be repeatedly melted and remelted to form a bond that is indistinguishable from the surrounding matrix. Anyway, the risk of polymer degradation is higher while in the case of the thermosetting matrix welding becomes more difficult (Messler, 2004).

Table 1 Suitable techniques for joining composite materials by type

<i>PMCs</i>	<i>MMCs</i>	<i>CMCs</i>
Adhesive bonding	Friction stir welding	Friction stir welding
Mechanical bonding	Brazing	Brazing
Fusion bonding (thermoplastic matrix)	Fusion welding	Fusion welding
Weld bonding (thermoplastic matrix)	Weld-brazing	Weld-brazing
Rivet bonding	Adhesive bonding	Brazing
	Mechanical bonding	Adhesive bonding

Joining Metal Matrix Composites

Metal matrix composites are widely diffused in some meaningful sectors (such as automotive and nautical), due to their capacity to merge the ductility and performances of metals and the characteristics of the reinforcement. They consist of a metal matrix with continuous reinforcement (Lynch and Kershaw, 2018). Metal-matrix composites can offer a variety of significant property advantages over unreinforced, monolithic metals and alloys, depending on the material and form of the reinforcement. The most diffused metal matrixes are Aluminum or aluminum alloys, but other metals like Magnesium and Titanium have also been used (Haghshenas, 2016; Kaczmar *et al.*, 2000). The reinforcements are usually in the form of short fibers; thus they don't show a strong anisotropy. Due to the metallic matrix, the main physical joining techniques (as welding, brazing, and FSW) are easily applicable for most of the materials employed. Mechanical joining is possible but discouraged due to the issues related to the stress field arising and the increment in terms of weight and of course, as with all the composites, the risk of fibers damage is high. Adhesive joints are rarely used since one of the main advantages of MMCS is its implementation for high-temperature applications (Messler, 2004). Anyway, the use of adhesive for MMCs can lead to interesting results as little to none surface preparation is needed and are suitable for thin metal sheets and for their low thermal insult to the materials bonded.

Joining Ceramic Matrix Composites

Among all the composite materials, the most difficult to join are the CMCs. Different from the other composites, CMCs usually consist of ceramic fibers (usually in the form of short fibers or whiskers) embedded in a ceramic matrix (Okada, 2003). The adding of reinforcement into a ceramic matrix aims to partially overcome the main issue of ceramic materials, namely their brittleness. Those composites are widely diffused in high-temperature applications such as gas turbines, heat exchangers, or turbine blades (Rosso, 2006). The joining method depends on reinforcement types. Due to their high brittleness, tensile stresses in the joint must be kept to an absolute minimum. For this reason, and for their hard behavior causing difficulty to make holes, the use of mechanical joints is highly discouraged (Bansal and Lamon, 2014). As for physical joints, the most suitable technique appears to be the FSW, as it does not require the melting of the parts. Anyway, brazing is the most feasible technique that offers the highest temperature serviceability, although portrays difficulties with the metallic braze filler alloy or with the thermal mismatch between the metallic filler and the ceramic matrix. As for chemical bonding techniques, most of the common adhesive used displays an insufficient toughness and are therefore not suitable, especially for the limited application of adhesive for high-temperature application.

Joining Dissimilar Joining

The joining of dissimilar materials has recently gained interest in the automotive and aerospace industries (Martinsen *et al.*, 2015). For example, joining aluminum alloys and polymers in the vehicle industry leads to a consistent decrease in costs during both the manufacturing process and during the lifetime of the vehicles. More care is needed when joining dissimilar materials. The most applied technique for joining dissimilar composites is adhesive bonding, being this technique suitable for most of the material used. The adhesive should be compatible with both parts. In the case of PMC to metals it is more difficult to prepare the mating surfaces by removing all traces of mold release agent, or another contaminant from the composite and grease or even oxide (especially for aluminum and titanium) on metal parts. However, in cases the adhesive bonding is not convenient or viable, mechanical bonding is preferred as an alternative. In the last years, polymer-filled concrete has received attention as tough, seismic vibration-tolerant masonry construction materials. In this last case, adhesive bonding using polymeric adhesives is preferred. Another well spread technique suitable for joining dissimilar materials is FSW technology, due to the lack of fusion of the involved materials. In particular, this technology can be applied to join different metal-based composites (Kah and Jukka Martikainen, 2013; Kumar *et al.*, 2015).

Conclusion

There are a lot of variables that influence the strength and the mechanical properties of composite joints, but their influence and implementation in industrial technologies still require further study. It is possible to join the typology of composites (namely PMC, MMC, CMC), but the suitable technique must be accurately chosen. Three primary methods of joining composites are discussed for each composite typology: mechanical fastening, adhesive bonding, and welding. Polymer-matrix composites or fiber-

reinforced plastics (FRPs) are usually adhesively bonded or mechanically joined by either fasteners or integral mechanical interlocking devices. Thermoplastic types can also be welded or thermally bonded. Metal-matrix composites, or MMCs, are the most versatile in joining and they can be brazed, adhesively bonded, or welded. In welding and brazing, the choice of the process or the filler must be considered in compatibility with the matrices. At last, CMCs display a wider number of issues and difficulties and further studies are still needed.

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Joining of Polymer Matrix Composites (PMCs) – Adhesive Bonding

Ricardo JC Carbas, Eduardo AS Marques, Catarina SP Borges, and Lucas FM da Silva, University of Porto, Porto, Portugal

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Nomenclature

b Joint width

CFRP Carbon Fiber Reinforced Polymer

k Bending moment factor

l Overlap length

M Bending moment

P Applied load

P_{AY} Maximum load which just creates adherend yielding

P_{GY} Failure load of the adhesive due to global yielding

SLJ Single Lap Joint

t_s Adherend thickness

σ_s Stress at the inner adherend surface

σ_t Normal tensile stress acting in the adherend

σ_y Yield strength of the adherend

τ_y Shear yield strength of the adhesive

Glossary

Adhesive failure A type of failure mode in adhesive joints, occurring when there is failure in the interface between the adhesive and the substrate.

Cohesive failure A type of failure mode in adhesive joints, which occurs when there is failure in the adhesive layer.

Composite joint A type of bonded joint where two composite substrates are joined by a substrate.

Delamination A type of failure mode in composite materials, occurring when there is a failure of the resin that fills the interlaminar space between the composite plies.

Peel stress Normal stresses that are generated in the transverse (through-thickness) direction of a composite material or adhesive layer.

Shear stress Stresses that are generated in a direction parallel to the cross section of the material or along the overlap length of an adhesive layer.

Single lap joint The most commonly used geometrical joint configuration, where two substrates overlap on one side.

Stress concentration Corresponds to a location where there is a peak in the stress distribution.

Transverse strength The strength of the material in the transverse (through-thickness) direction.

Introduction

The main advantage associated with the now widespread use of structural adhesives is the fact that they are able to effectively join dissimilar materials, something which cannot be easily performed with other conventionally used joining methods. Adhesive bonding is particularly interesting for use with composite materials, when compared with classical joining methods, as it does not require holes to be drilled, avoiding problems related to the fact that composites are highly sensitive to notches. Within the design of composite structures, adhesive bonding is also extensively used to bond metallic fittings or components to these structures.

Composite materials combine dissimilar constituents in order to obtain a new material, with better properties than that of each component used individually, providing high strength with lower weight, when compared, for example, with metallic alloys. These materials are known for their extremely high specific strength and stiffness, excellent corrosion resistance, good thermal stability and high fatigue resistance. Due to these very advantageous properties, composite materials are widely used in aeronautical, aerospace, automotive and marine industries.

The mechanical behavior of most composite materials is known to be highly anisotropic, with regards to both the stiffness and strength. While composites exhibit excellent properties in the fiber direction, the opposite occurs in the transverse direction, where the stiffness and strength are in fact much lower. When bonding composite materials, the main problem that arises is related to the low transverse tensile strength (peel strength), which is of the same order of magnitude or lower than that of the matrix and can lead to delamination failure.

The single lap joint (SLJ) is a geometrical adhesive configuration that is commonly found in many industrial applications, mainly due to its simplicity and efficiency. However, the main problem associated to this joint is the fact a non-uniform stress distribution (shear and peel) is generated along the overlap length. In fact, a stress concentration arises at the ends of the overlap, as shown in Fig. 1.

It is this peak peel stress, at the ends of the SLJ overlap length, that can induce composite failure, as it loads the composite in the transverse direction, which has low transverse tensile strength. The composite adherend splits locally due these peel stresses and the shear transfer capacity between both adherends is destroyed, promoting the premature failure of the composite joints (da Silva *et al.*, 2011).

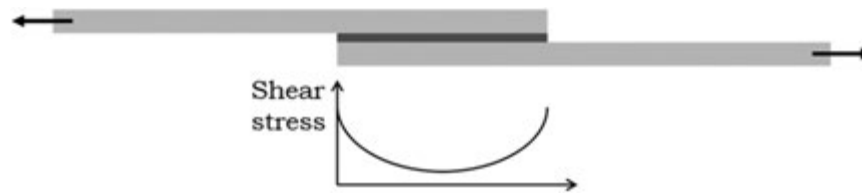


Fig. 1 Single lap joint configuration and shear stress distribution along the overlap length.

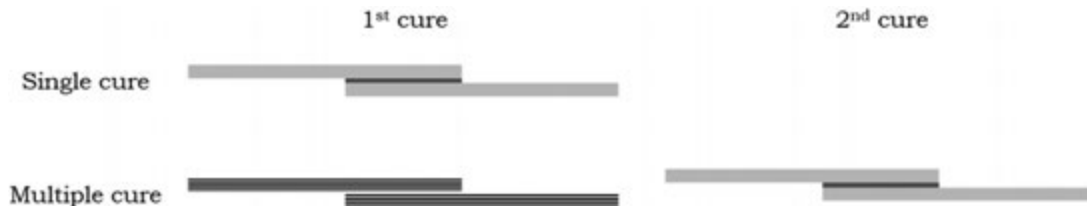


Fig. 2 Schematic of the common manufacturing bonding processes between composite components.

Factors that Affect Joint Strength

The strength of a bonded joint is known to depend on a large number of parameters and can be significantly improved with an appropriate selection of the manufacturing process, adequate surface preparation of the adherends, optimized geometrical characteristics (such as adhesive thickness or overlap length), and correct selection of the adhesive type.

Manufacturing Process

The manufacturing process is an extremely important aspect that needs to be taken into account during the design of an adhesively bonded joint. Although the manufacturing process can have significant implications on the economic viability of a component with bonded construction, it can also have an important effect on the mechanical performance of the joint, as both the failure mode and joint strength can be influenced by the manufacturing process (Budhe *et al.*, 2017; Shang *et al.*, 2019b; Park *et al.*, 2010; Markatos *et al.*, 2013; Mohan *et al.*, 2014a; Arenas *et al.*, 2013).

The manufacturing process of composite joints can be divided into two main groups, where the manufacture is either performed in a single-cure or in a multi-cure instead (see Fig. 2). In a single cure manufacture, the co-bonding process (where only one adherend is cured with the adhesive) or the co-curing process (where both adherends are cured with the adhesive) are included. For the multi-cure techniques, the adherends are completely cured individually and only after secondary cure of the adhesive is performed the complete cure of the adhesive joint is achieved. The main goal of using the single cure is to save time. However, an additional issue arises as the adhesive must be fully compatible with the composite resin. Compatibility must be ensured with regards to the time and temperature of the cure process but also with regards to the chemical compatibility of both the adhesive and the composite matrix resin, which will be mixed during the curing process. For complex adhesive joints, the use of multi-cure techniques can be a useful step to simplify the bonding process.

Some studies can be found in literature, comparing the performance of joints cured in single cure or multi-cure process, which indicate that the single-cure provides lower strength than the joints cured in more than one cycle of cure (when the adherends and adhesive are cured individually). This decrease can be explained mainly by varying degrees of chemical incompatibility between the adhesive and the composite resins, but also by the level of absorbed moisture present in prepreg. This moisture will be released as a vapor during the curing process, creating a weak adhesion between the adhesive and composite adherend (Mohan *et al.*, 2014a, 2015; Song *et al.*, 2010; Mohan *et al.*, 2013).

Surface Preparation

Surface treatment has an important role in the performance of adhesive joints, as the joint strength can be negatively affected by the presence of surface contaminants and the low chemical or physical compatibility between substrates and adhesives. The main purpose of surface treatment is, therefore, to provide a substrate surface without contaminants, high surface energy and chemically activated of surface, which will, in the short term, ensure good adhesion and, in the long term, ensure durability and stable mechanical performance of the joint (da Silva *et al.*, 2009; Kanerva and Saarela, 2013; Islam *et al.*, 2014; Encinas *et al.*, 2014; Sinmazçelik *et al.*, 2011). The selection of a correct surface preparation must not only provide high joint performance immediately after manufacture, but also ensure that this level of strength is maintained over the service life of the bonded component, without premature failure of the adhesive joints.

The phenomenon of adhesion between an adhesive and adherends is complex and can rely on a combination of chemical, physical and mechanical interactions, which must be stable to ensure durability (Encinas *et al.*, 2014; Borsellino *et al.*, 2009; Palmieri *et al.*, 2016; Mohan *et al.*, 2014b). Several surface treatments can improve adhesion by enhancing the level of mechanical interlocking between the adhesive and the substrate. Among these methods are the use of grit blasting, abrasion using emery paper and the use of a peel ply. The application of some of these methods, however, requires special care, as grit blasting and abrasion with emery paper do have a high risk of damaging the fibers. In addition, the large quantity of residues generated with the use of these methods requires a subsequent thorough cleaning with solvents to avoid contamination of the adhesive and the interface. In contrast, the use of a peel ply reduces the risk of fiber damage, consisting in the insertion of protective sheets in the outer surface of the composites prior to curing. This is a clean and effective method to improve the adhesion of bonded surfaces and it is very commonly used in the aerospace industry, although it slightly increases the complexity of the manufacturing process by introducing an additional step. However, this method must be used carefully, as the material of the peel ply can interact with the resin of composite or the adhesive during cure at high temperatures. The selection of peel ply needs to take into account the chemical composition of the peel ply and ensure chemical compatibility between the resins and adhesive. Due to this fact, a few studies have suggested that the strength or fracture toughness of joints obtained using peel ply is not higher than that exhibited by joints where the surfaces were simply treated mechanically (abrasion techniques) (Encinas *et al.*, 2014; Kreling *et al.*, 2013; Fischer *et al.*, 2012; Kanerva *et al.*, 2015).

The surface treatment methods used to improve the chemical interaction usually rely on the introduction of oxygen species on the surface of the adherends, which will react with adhesives that are rather hydrophilic and exhibit chemical functionalities. For composite materials this type of chemical modification can be easily achieved with the use of a flaming, plasma or laser treatment. Despite being mainly chemical in nature, these techniques can also induce changes in the surface morphology that will also provide a mechanically driven improvement of the adhesion level (an interlocking effect). In order to increase the durability of adhesively bonded joints with composite substrates and enhance their fatigue life resistance, it is therefore crucial to select a suitable surface treatment that is able to provide the necessary chemical compatibility between adhesive and matrix resin as well as ensuring a suitable level of surface roughness (Dawood and Rizkalla, 2010; Azari *et al.*, 2010; Heshmati *et al.*, 2015; Rechner *et al.*, 2010; Brack and Rider, 2014). Prolongo *et al.* (2009) evaluated the strength, wettability and adhesion performance of composite joints with different surface treatments (peel ply, grit blasting and plasma). It was shown that the surface treatment used has influence on the failure mode and in the fracture mechanisms. In this work, the plasma treatment was found to provide the highest strength and peel ply was the treatment that provided the lowest values.

Geometrical Characteristics

The strength and failure mode of composite joints, as well as any other type of adhesive joint, is known to be highly dependent on the geometrical characteristics of the joint. Several geometrical parameters, such as the adhesive thickness and the overlap length have been extensively studied by many authors and its influence on the mechanical behavior of the joint is described in this section.

Adhesive thickness

The thickness of the adhesive layer is an important geometrical characteristic that needs to be carefully controlled during joint manufacture as, in order to reach the best joint performance, an optimum thickness for the adhesive must be determined and implemented. For the adhesives which are supplied in paste form, this can be achieved by controlling the pressure applied as a function of the adhesive viscosity and using spacing elements, such as beads and shims. For adhesives that are supplied in a film form, the thickness can be controlled with the use of carrier layers, embedded within the film itself. Most structural adhesives exhibit optimal mechanical behavior for thicknesses between 0.1 and 0.2 mm, with the literature describing a gradual strength decrease as the thickness increases above this range. Several explanations have been proposed for this decrease in joint strength, such as the higher susceptibility to the appearance of defects, for example air bubbles and microcracks, in larger, thicker adhesive layers. In addition, in a SLJ, higher adhesive thickness increases the bending moment at the ends of the overlap, creating larger peel and shear stresses that can overload the adhesive and/or substrates, resulting in a decrease of joint strength (da Silva *et al.*, 2011).

The work of Nascimento (2013) presents a relevant study on the influence of the adhesive thickness on the mechanical behavior of SLJs with CFRP substrates, bonded using an adhesive with intermediate ductility. This work has shown that as the adhesive thickness increases, the joint strength can gradually decrease up to a point where it only supports 50% of the initial failure load.

Overlap length

The correct selection of an overlap length is a key factor for maximizing the performance of bonded joints with composite substrates, as non-optimal values of overlap length will lead to premature failure of the adhesive and substrate without extracting the full potential of these materials.

Literature shows that for a SLJ configuration it is possible to define a certain value of overlap length above which there is no strength advantage to be gained by increasing the overlap. A plateau in joint strength is indeed reached and any further increases in the overlap length will lead to a highly inefficient joint, that is, one that uses excessive material for achieving a given performance

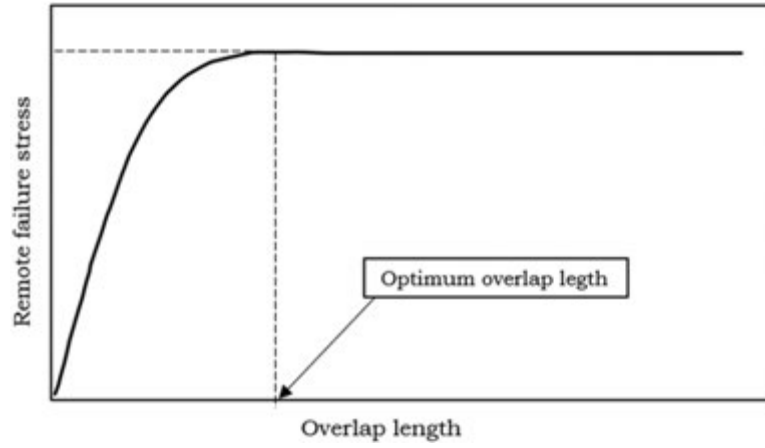


Fig. 3 Repair strength as function of the overlap length.

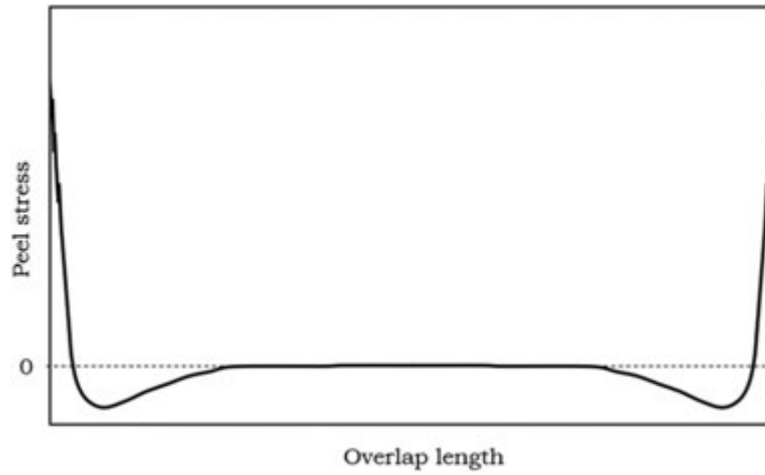


Fig. 4 Peel stress distribution in a single-lap joint as function of overlap length in the elastic region.

target. This phenomenon occurs because the central region of the overlap will eventually become unloaded (unused to transfer load), as the stress is concentrated at the overlap ends. More critically, when composite adherends are used, the strength of the joint will eventually become restricted by the strength of the composite matrix, as the transverse loads generated at the overlap length will eventually exceed the transverse strength of the composite. This can be seen in Figs. 3 and 4. If a sufficiently strong adhesive is used, the maximum efficiency in joint strength is obtained when the substrate fails.

In order to simplify the design process of adhesive joints (in practice, the material selection and the geometrical dimensioning of the joint) Adams *et al.* (1997) proposed a simple methodology to determine the overlap and the adherend thickness for single lap joints (see Fig. 5). The load corresponding to the total plastic deformation of the adhesive (global yielding) is

$$P_{GY} = \tau_y b l \quad (1)$$

where P_{GY} is the failure load of the adhesive due to global yielding, τ_y is the shear yield strength of the adhesive, b is the joint width and l is the overlap length. The normal tensile stress (σ_t) acting in the adherend due to the applied load P and is given by

$$\sigma_t = P / b t_s \quad (2)$$

where t_s is the adherend thickness. The stress at the inner adherend surface (σ_s) due to the bending moment M is given by

$$\sigma_s = 6M / b t_s^2 \quad (3)$$

where $M = k P t_s / 2$, according to Goland and Reissner (1944). The variable k is the bending moment factor which reduces as the lap rotates under load. The maximum load that can be carried which just creates adherend yield (P_{AY})

$$P_{AY} = \sigma_y b t_s / (1 + 3k) \quad (4)$$

where σ_y is the yield strength of the adherend. For low loads and short overlaps, k is approximately 1. Therefore, for such a case

$$P_{AY} = \sigma_y b t_s / 4 \quad (5)$$

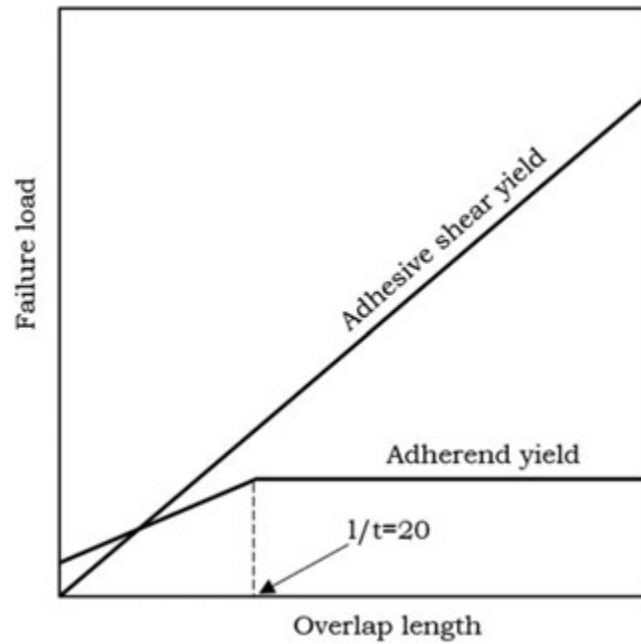


Fig. 5 Methodology to predict the overlap corresponding to adherend yield.

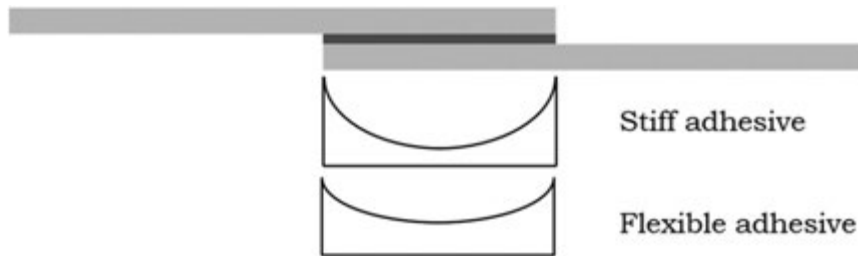


Fig. 6 Stress distribution along the overlap for brittle and flexible adhesives.

However, for joints which have overlaps significantly longer than the adherend thickness, defined as having an $l/t_s \geq 20$, the value of k decreases and it is assumed that tends to zero. In this case, the whole of the cross-section yields is

$$P_{AY} = \sigma_y b t_s \quad (6)$$

From a certain value of $l/t_s > 20$, the strength increase is marginal and the total weight of the joint and its dimensions are increased unnecessarily (Campilho *et al.*, 2005; Da Silva and Adams, 2007b). It is important to state, however, that this ratio between joint strength as a function of overlap length is also highly dependent of adhesive behavior, as discussed in the Section “Type of Adhesive”.

Type of Adhesive

Joint design efficiency can be assessed by considering the stress distribution of a simple lap shear or peel joint. It will become evident that the stress distribution of the joint is highly dependent on the adhesive behavior. Generally, joints with flexible adhesives will exhibit a more uniform stress distribution, while the use of brittle adhesives will impose higher stress concentrations at the ends of overlap length (see Fig. 6).

Flexible adhesives exhibit high toughness, with good resistance to crack propagation and can withstand peel stresses, combined with excellent performance under fatigue and impact loads. Due to their viscoelastic and viscoplastic behavior, ductile adhesives show a large degree of strain rate dependence, with their mechanical behavior changing significantly as a function of strain rate. Ductile adhesives are less affected by the stress concentrations generated in bonded assemblies and are capable of joining materials with very different thermal expansion coefficients. Typically, the joint strength increases up to the adhesive allowable ductility. The ductile behavior of the adhesives provides an increased capacity to withstand peak loads and shows large damage tolerance to flaws and defects (such as voids and porosity) in the bondline. In contrast, brittle adhesives are stronger and stiffer but have markedly lower toughness. Brittle adhesives are more densely cross-linked and are especially well suited to use in structural applications that

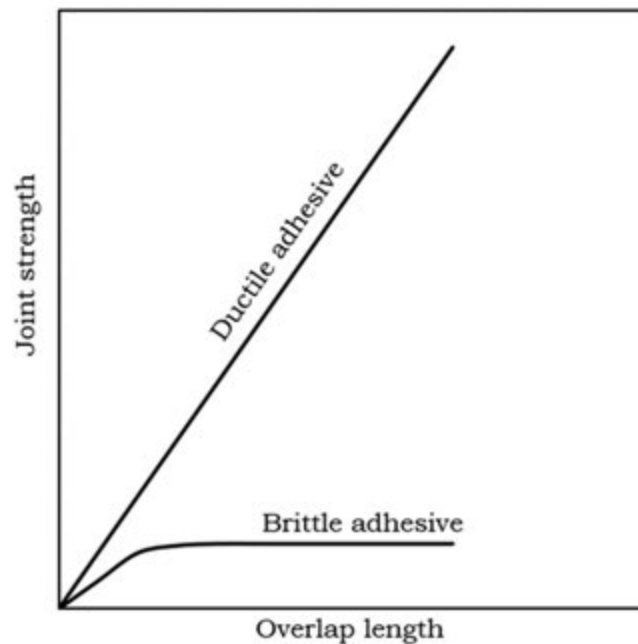


Fig. 7 Influence of joint size on selection of joint configuration. Adapted from Shang, X., Marques, E., Machado, J., *et al.*, 2019b. Review on techniques to improve the strength of adhesive joints with composite adherends. *Composites Part B: Engineering* 177, 107363.

must resist high temperatures and aggressive environmental conditions. However, due to their stiffness, they are prone to generate very high stress concentrations which can cause delamination in composite adherends with low transverse strength. The design of adhesively bonded composite joints exclusively using brittle adhesives (adhesives purely elastic) limits the joint capabilities, precluding the use of substrates of dissimilar materials or with highly distinct coefficients of thermal expansion.

As stated above, the effect of the overlap length is highly dependent on the type of adhesive. Considering a case where the adherends do not suffer plastic yielding (remaining in the elastic phase) and that the adhesive used is highly ductile (e.g., shows a shear strain to failure higher than 20%) the joint strength is approximately proportional to the overlap length, as the adhesive will deform plastically and uniformly along the whole overlap. In contrast, if the same adherends (that remain in elastic domain) are used and the adhesive is replaced by another with a brittle behavior, the joint strength will not be proportional to the overlap length because the stress (and the plastic deformation) is now almost fully concentrated at the ends of the overlap. In Fig. 7, a schematic representation of joint strength as a function of overlap length used can be seen, for both types of adhesive behavior (brittle or ductile behavior).

In literature, several studies that evaluate the joint strength performance of SLJ as a function of overlap length for different adhesives are available (Li *et al.*, 2015; Neto *et al.*, 2012). Three different adhesives were used: a brittle epoxy adhesive (Araldite® AV138), an intermediate stiffness epoxy (Araldite® 2015) and a ductile polyurethane (SikaForce® 7888). In Fig. 8, it can be noticed that the strength of joints bonded with the brittle adhesive increases until a plateau is reached and from this point further the failure load was controlled by the composite. The joints bonded with brittle adhesive showed a smaller improvement of failure load with increase of overlap length due to limited plastic deformation of the adhesive. Typically, when the stress concentration at the ends of the overlap reaches the maximum strength of the adhesive, the crack start to propagate, leading to premature failure. For the joints bonded with the intermediate and the ductile adhesive, linear behavior can be noticed as the failure was completely cohesive in the adhesive.

During the tensile test, plastic yielding of the adhesive takes place at the ends of overlap where the stresses are concentrated, together with the redistribution of stresses in the adhesive layer towards the inner overlap regions (McGeorge, 2010; Davis and Bond, 1999). Consequently, a nearly proportional relationship can be seen between the failure load and overlap length (see Fig. 8). This proportional behavior is more pronounced for the joints with ductile adhesive, due to the larger yielding of the adhesive.

For ductile adhesives, with a more plastic dominated behavior, the remote failure stress increases proportionally with the overlap length and the failure occurs in the adhesive. But the failure mechanism itself can change and delamination of a composite adherend can occur if the peel strength of adhesive is higher than the transverse strength of the composite material. For example, when SLJs are loaded, the bending moment at the ends of the overlap promotes a higher level of peel load which will translate into a high level of peel stress promoting adherend failure between the plies (delamination).

Methods to Increase Joint Strength

A wide variety of different techniques has been proposed to increase the strength of composite adhesively bonded joints, such as the use of adhesive fillet, mixed-adhesives, Z-pins, stitching, fiber metal laminates or adherends toughening. The working principle

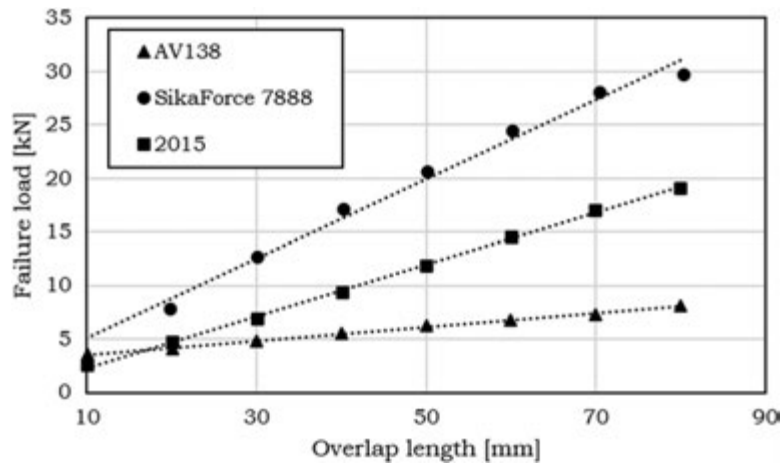


Fig. 8 Failure load for three adhesives as a function of overlap length.

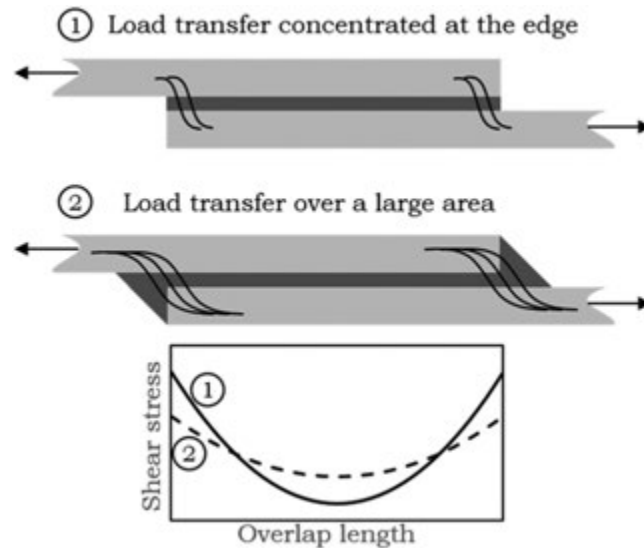


Fig. 9 Load transfer and shear stress distribution in single lap joints without and with fillet. Adapted from Da Silva, L.F.M., Öchsner, A., 2008. Modeling of Adhesively Bonded Joints. Springer.

behind most of these methods is to introduce a reinforcement on the composite and increase the transverse strength of the composite adherends. This will prevent early failure by delamination and make full use of the mechanical properties of both the composite and the adhesive. The following sections describe in greater detail the most important of these methods.

Adhesive Fillet

The load transfer and shear stress distribution of a single-lap joint with and without a spew fillet are schematically represented in Fig. 9. The stress concentration for the single lap joint with a square end is concentrated locally at the ends of the overlap length. The spew fillet (spews are the excess of adhesive squeezed out of the lap region at the moment of the joint manufacture) effectively increases the load transfer area and smoothens the sharp angle present in this region (Da Silva and Öchsner, 2008).

The use of spew fillet is interesting as it is purely geometrical modification of adhesive joints and alters the stress intensity factors of the joint, providing a significant reduction of peel and shear peak stresses and leads to an increase of joint strength. This joint strength increase is highly dependent on the fillet shape and the mechanical characteristics of the adhesive (da Silva *et al.*, 2011; Tsai and Morton, 1995; Lang and Mallick, 1998; Rispler *et al.*, 2000; Belingardi *et al.*, 2002).

The stress intensity factors in adhesive lap joints change greatly according to the shape of the fillet. Fig. 10 shows schematically that the weak point present in the joint without fillet is the point where crack propagation will start, as the stress concentration is located in a single point. In contrast, for the lap joints with spew fillet there is no single point with a high level of stress concentration, providing a stress distribution in a large area, promoting a decrease of peel stress.

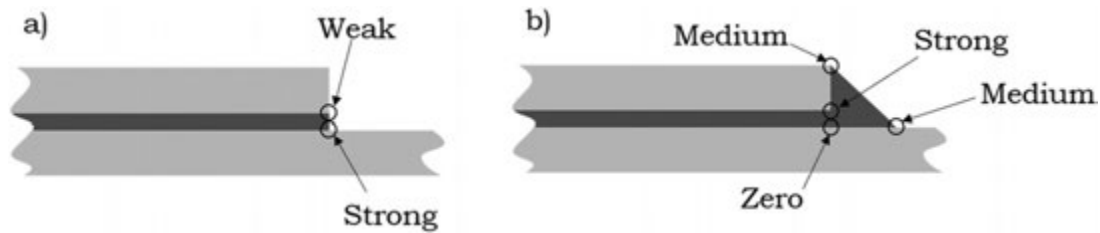


Fig. 10 Location and intensity of stress concentrations on adhesively bonded joints (a) without and (b) with spew fillet geometry. Adapted from da Silva, L.F., Öchsner, A., Adams, R.D., 2011. Handbook of Adhesion Technology. Springer Science & Business Media.

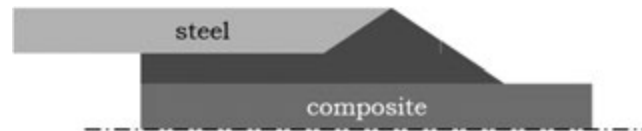


Fig. 11 Internal taper and adhesive fillet.

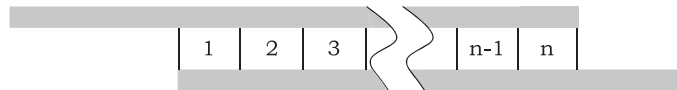


Fig. 12 Mixed adhesives bondline.

Adams and Mallick (1992) and Da Silva and Adams (2007b) achieved a reduction of the transverse tensile stresses in the composite adherend, as well as an increase of the joint strength using an internal tape and adhesive fillet (see Fig. 11). The use of this technique was able to change the failure mode as delamination was avoided. While this is a very powerful technique to increase the joint strength and prevent the delamination, its suitability for practical application is low, as its manufacture process requires costly and time-consuming additional machining or forming operations.

Adhesive Stiffness Modification

The introduction of variations on the mechanical properties of an adhesive along the overlap length of a bonded joint serve as the basis for a set of techniques that allow improvements of the stress distribution and an increase of joint strength. Their underlying concept relies on establishing a gradual modification of the adhesive stiffness, increasing joint strength by minimizing the large stress concentrations at the ends of the overlap length, first proposed by Raphael *et al.* Raphael (1966) (see Fig. 12). Some of the techniques that rely on the modification of the adhesive stiffness are the use of mixed adhesives, the physical modification of the adhesive and the adoption of functionally graded adhesives.

Mixed adhesives

Mixed adhesive joints are those which use two (or more) discrete adhesives in the same overlap, usually selected to present highly distinct mechanical properties. In a two adhesive mixed adhesive joint, a stronger, yet brittle, adhesive is typically used in the middle of the overlap while a more flexible, yet weaker, adhesive is placed at the ends of the overlap length. Hart-Smith (1973) was the first author to propose and analytically study this technique, demonstrating that an effective joint strength increase could be attained. The use of a flexible adhesive at the ends of the bondline length allows for the reduction of peel stress at the ends of the overlap length, obtaining a more uniform shear stress distribution along the bondline, reducing the stress concentration and consequently preventing the delamination of composite adherends. Through this technique, the shear stress is greatly improved but points of high stress concentration still occur at the interfaces that separate the different adhesives (see Fig. 13). Nonetheless, the mixed adhesive technique provides joint strength improvements in comparison with joints with the adhesive used individually (brittle or flexible adhesives) (Da Silva and Adams, 2007b; Pires *et al.*, 2003; Temiz, 2006; Bouiadjra *et al.*, 2007; Fitton and Broughton, 2005; Da Silva and Lopes, 2009; Da Silva and Adams, 2007a; Machado *et al.*, 2018). Machado *et al.* (2018) showed that with this technique it is possible to increase the joint strength up to 64% when compared to the individual adhesive joints. The main problem associated to the mixed adhesive technique is the necessity of introducing a physical separation barrier of adhesive between bands. Marques and da Silva (2008) proposed a separation of the adhesive through thin silicone strips, which have the added benefit of being able to control the adhesive thickness.

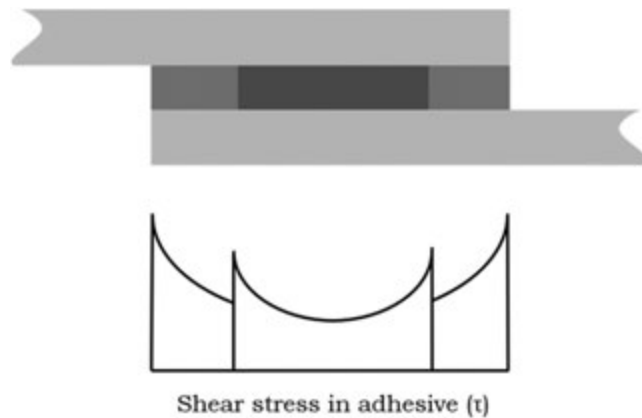


Fig. 13 Mixed adhesives technique and shear stress distribution.



Fig. 14 Graded adhesive joint using particles dispersed in the adhesive.



Fig. 15 Schematic concept of the functionally graded joints obtained by induction heating.

Physical modification of the adhesive

The process of physically modifying the adhesive is a technique based on the mixed-adhesive concept, which uses physical modification to obtain different properties in different locations along the overlap length. The physical modification can be simply achieved by adding particles to the adhesive, for example, rubber particles. Many other types of toughening particles are also available, including nanomaterials, such as single and multi-walled carbon nanotubes, carbon black nanoparticles, thermally expandable particles, clays and various oxides (see Fig. 14).

The use of tough particles (such as rubber) is an effective method to reinforce the adhesive at the ends of overlap length, improving the adhesive's fracture toughness locally. Due to the low elastic modulus of rubber, when the crack reaches a rubber particle the local stress concentration at the crack tip is reduced, stopping or at least slowing the crack growth rate (Sancaktar and Kumar, 2000; Kinloch and Taylor, 2002). The particle size and dispersion pattern are crucial factors in this technique if joint performance improvements are the final goal (Bagheri and Pearson, 2000; Stapleton *et al.*, 2012). Some particles, such as carbon black, nanotubes, and thermally-expandable particles have a dual role, providing not only an increase of joint strength but also other properties such as increased conductivity (Carbas *et al.*, 2017) and self-dismantling capabilities (Banea *et al.*, 2017). The main challenge associated with this approach is the process used to distribute the particles in an accurate and repeatable way along the bondline. One solution for achieving this objective is to use short bands of adhesive with different amounts of particles (Carbas *et al.*, 2017) or to use a process that is able to move the particles along the bondline towards pre-set positions (Marques *et al.*, 2019). This technique uses a custom designed magnetic field that acts upon magnetized particles to vary the particle distribution along the bondline (da Silva *et al.*, 2019). The main limitation of this process is the necessity of using magnetic particles or subjecting them to a coating process with a magnetic material.

Functionally graded adhesive

The mixed-adhesive and the physical modification techniques can be considered as a rough version of a true functionally graded adhesive. The ultimate concept is to obtain a functionally modified adhesive, with properties that vary gradually (non-step-wise) along the overlap (see Fig. 15), allowing for a true uniform stress distribution and avoiding points of high stress concentration that occur at the borders between different adhesives (as present in mixed-mode technique).

The concept of functionally graded adhesives requires adjusting the distribution of the mechanical properties along the adhesive layer to ensure maximum performance, which can be a highly complex process given the large range of material properties that can be considered and varied gradually. A few analytical models were developed to optimize the graded mechanical properties and the shape of the distribution of the mechanical properties along the bondline. Models exist for tubular,

SLJs, L-joints, T-joints and reinforced patches with a functionally graded bondline (Kumar, 2009; Kumar and Pandey, 2010; Kumar and Scanlan, 2013; Carbas *et al.*, 2013; Stein *et al.*, 2016). These simple models are used to evaluate the stress distribution and predict the failure load of the functionally graded joints.

The work of Carbas *et al.* (2014a) proposes an apparatus to provide a differential cure of bonded joints using induction heating technology. Using special induction coils to control the temperature applied to different areas of the adhesive, the adhesive stiffness was successfully varied gradually along the overlap, being maximum in the middle and minimum at the ends of the overlap. When testing the joint, an increase of more than 60% in failure load was obtained for the graded joint, compared to joints with an homogeneously cured bondline (Carbas *et al.*, 2014a). The same apparatus was used to perform repair on wood beams with CFRP patches, again leading to joint strength improvements, with the bonded beams repaired with graded adhesive layers performing better than repaired beams which were bonded with an adhesive with uniform properties along the bondline (Carbas *et al.*, 2014b).

The main limitation of these graded joints obtained by differential heating is the fact that the adhesive properties can be easily modified if the joint is submitted to temperatures above the T_g of the adhesive. In this case, the temperature completely reverses the grading process and creates a joint with uniform properties, the same as those of a joint cured isothermally (Carbas *et al.*, 2015). This is because the technique changes the properties by reaching different levels of cross-linking at different temperatures and, when submitted to temperatures above the T_g , the level of cross-linking of the adhesive is uniform along the adhesive layer, leading to uniform mechanical properties. An alternative technique that maintains the functionally graded properties for long term is to use the honeymoon technique, able to provide a stable functionally graded variation of the properties as a function of temperature (Kawasaki *et al.*, 2016). This technique uses two different adhesives, both mixed with different amounts of hardening compound using a fully automated application machine, providing graded mechanical properties, stable in the long term. The main limitation of this technique is the fact that very specific adhesive formulations must be employed to ensure that a large degree of variation of mechanical properties can be attained.

Adherend Stiffness Modification

Another approach to prevent composite delamination is to change the transverse stiffness of the adherends. The stiffness of the adherend can be significantly modified through the thickness direction, reinforcing the composite with metal or polymeric layers. The inclusion of additional layers with a different stiffness will change the peel strength of composite. The stress concentration (mainly composed of peel stresses) is higher at the ends of the overlap length and if these additional layers are placed on the surface of adherend a reduction of the peel stress can be attained. When the peel stress is transferred to these layers, they are able to redistribute it along a larger area (Carbas *et al.*, 2015).

Tougher adherend reinforced by metal

The most commonly used technique to increase the composite transverse strength is to include metal layers within the composite layup (see Fig. 16). This concept was developed based on the fiber metal laminate (FML) materials but, while FMLs use composites to reinforce a component which is mainly metallic in nature, the main purpose of a hybrid laminate is to reinforce a mainly composite material with the addition of metal layers. As is the case for the FML, the hybrid laminate concept combines the properties of composite materials and metals such as high strength, low density, fatigue resistance and increased tolerance to impact damage. By increasing the transverse strength, premature failure of the adherend is avoided which makes this materials very suitable for use in aeronautical structures (Palmieri *et al.*, 2016; Vogelesang and Vlot, 2000). The main challenge associated with this approach is to ensure good adhesion between the fiber and metal interface, essential to ensure that the stress is fully transferred to the reinforcing material. Recent works have studied several design and manufacturing aspects of the hybrid laminates, considering factors such as the use of different materials and surface preparations as well as the inclusion of an additional adhesive inter-ply layer between the composite-metal interface (Hader-Kregl *et al.*, 2018; dos Santos *et al.*, 2019). The authors found that optimal joint performance is achieved when the metal is chemically treated (using a method such as anodization), showing the highest peel strength. Moreover, it was found that the joint strength can be further increased, if an additional adhesive layer, is used on the composite-metal interfaces, although this solution greatly increases cost and complexity.

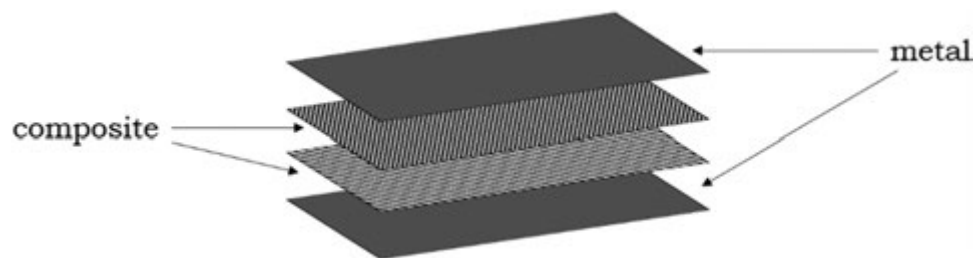


Fig. 16 Schematic representation of hybrid laminate.

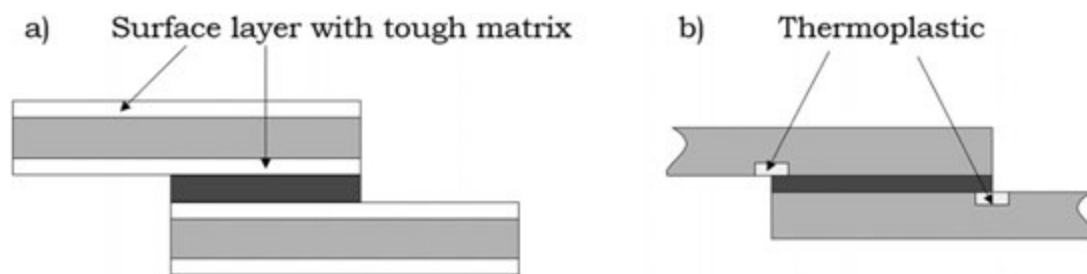


Fig. 17 Schematic of surface toughening techniques. Adapted from Park, Y.-B., Song, M.-G., Kim, J.-J., Kweon, J.-H., Choi, J.-H., 2010. Strength of carbon/epoxy composite single-lap bonded joints in various environmental conditions. *Composite Structures* 92 (9), 2173–2180. Schollerer, M., Kosmann, J., Völkerink, O., Holzrüter, D., Hühne, C., 2019. Surface toughening – A concept to decrease stress peaks in bonded joints. *The Journal of Adhesion* 95 (5–7), 495–514.

Other works have studied the effect of different hybrid laminate geometrical configurations on joint performance, showing that the hybrid laminates with metallic layers on the surface are able to avoid adherend delamination, preventing premature joint failure and consequently increase the joint strength (Palmares, 2016; Bano *et al.*, 2017). Recently, the joint performance of hybrid laminates joints was evaluated under different loading rates and an increase of joint strength was obtained with the increase of the loading rate (dos Santos Pereira, 2019). The volume ratio of the composite and metal is another parameter that needs to be considered, especially for aerospace applications, as it drastically affects the weight of the joints and also its delamination prevention capabilities. dos Santos Pereira (2019) showed that a ratio of 50:50 (assessed between the thickness of composite and that of the metal) provides good joint performance both for low and high loading rates, being optimal for use with adhesives which possess intermediate peel strength or high strength.

Tougher adherend reinforced by polymers

Another method which has been recently proposed to increase the transverse strength of composite material is the reinforcement of the composite materials with high toughness polymeric layers, much in the same way as metallic sheets are added. The reinforcement with a polymeric layer can be performed globally or just locally, that is, concentrated on the area where the peel stresses are higher. The global reinforcement with tough polymeric layer technique was proposed by Shang *et al.* (2019a) and improves the delamination resistance of a CFRP by including a toughened surface layer composed of a high toughness, crash resistant hybrid epoxy resin. The use of tough composite adherends has been found to avoid or at least delay the onset of delamination in composite adhesive joints (dos Santos Pereira, 2019; Shang *et al.*, 2019a). A schematic representation of this concept is shown in Fig. 17(a), where the high toughness resin (e.g., tough adhesive) is co-bonded on the both surfaces of composite material (the composite material most used to reinforce with tough external layers is the CFRP). To enhance strength and energy absorption, the outer layers of tough resin can be reinforced with a woven fiber matt. This technique allows to obtain joint strength improvements of more than 25% when compared with joints specimens manufactured using CFRP only. The joints were tested at different loading rates and an increase of joint strength performance was reached. It was also reported that in order to avoid the delamination of the composite it is necessary to ensure a minimum thickness of the tough layer, otherwise the severity of the peel strength transferred to the composite fiber during the test is still sufficient to induce delamination (dos Santos Pereira, 2019).

The local reinforcement with a tough polymeric layer technique (see Fig. 17(b)) was first proposed by Schollerer *et al.* (2019). This localized technique consists in inclusion of thin bands of thermoplastic at the ends of the overlap length. This corresponds to the location of the joint that will be subjected to high level of local stress which will be transferred as a peel stress to the composite adherend. Much in the same way as described for the use of the toughened surface layer, by adding these thermoplastic bands, it is possible to redistribute this stress and mitigate the load transferred to the composite. The main limitation associated with the insertion of thermoplastic bands is the danger of introducing misalignments and noncontinuity of the fibers that are located at the interface between adherend and adhesive. The volume fraction of the material that surrounds the bands is also different of the rest of composite (being rich in resin) and this can introduce another weak point.

The surface toughening techniques (applied globally or locally) can therefore be used to manufacture laminates with higher load transferring capability in the transverse direction (through the thickness) and that can be achieved changing the toughness of the polymeric layer and the thickness of the reinforcement materials used. When tough surfaces are used in laminate adherends the damage resistance increases, avoiding the premature composite adhesive joints failure and increase the length of the joint's life cycle.

Stiffness Joint Reinforcement

Full joint stiffness modification is another method that has been proposed to increase the joint strength and prevent its delamination. This improvement can be achieved by the insertion of discrete reinforcement elements in the transverse direction (through the thickness), crossing both the adherend and the bondline. The techniques based on this concept can be divided into those that use metallic inserts and those that use fibers for this purpose.



Fig. 18 Delamination prevention methods: Z-pin.



Fig. 19 Delamination prevention methods: stitching.

Transverse pins insertion

The transverse pins insertion technique, typically referred as the z-pins technique, consists in the insertion of small pins (of fibrous or metallic nature) through the thickness direction of the composite, as shown in [Fig. 10](#). These pins, which are inserted while the matrix is uncured, improve the peel strength of a composite material because this solution is able to combine the strength of the resin of composite with the strength of the pins. In addition, some strength is also provided by the friction generated between the pins and the composite layers ([Mouritz, 2007](#)). This technique ensures a transverse strength improvement, maintaining the laminate fiber layers together, and a reduction of premature damage (such as delamination).

[Pingkarawat and Mouritz \(2014\)](#) and [Mouritz \(2007\)](#) performed a review of Z-pin techniques used composite laminates and showed that this technique allows to improve the peel strength of composite materials, preventing the delamination. These authors have also highlighted the importance of this solution aeronautical and automotive applications, citing its effectiveness and low added weight ([Figs. 18](#) and [19](#)).

This technique has been shown to be highly effective in increasing the transverse strength and fracture energy of composite materials, with these mechanical properties increasing with the number of pins used ([Nguyen et al., 2016, 2017](#); [Koh et al., 2012](#); [Beylergil et al., 2011](#); [Arnautov et al., 2015](#)). However, these improvements are reliant on a suitable adherend geometry (for example, in thick adherends this technique is more effective) ([Li et al., 2016](#)) and also on the nature of the pins (as carbon fiber pins provide more significant improvements ([Ravindran et al., 2017](#))). The Z-pin technique was also adapted to use Z-pins anchored to metallic adherends. This anchoring can be achieved by a machining or welding process, using perpendicular or angled pins with flattened ends at the surfaces of the composite adherend, thus increasing pull-out strength ([Arenas et al., 2013](#)). This technique can also be used in dissimilar joints (those using different substrates being bonded together), where the differences in geometry create unbalanced stresses that can increase unwanted peel loads ([Nguyen et al., 2017](#); [Ucsnik et al., 2010](#); [Parkes et al., 2014](#)). Using these anchored Z-pins it is possible to increase the joint strength by 6.5 times when compared to unpinned joints, the elongation by more than 4 times and achieve energy absorption capacity 80 times higher ([Parkes et al., 2014](#)). Other novel design concept is the insertion of thin metal sheets between co-bonded laminates at the location of high peel stress levels (such as at the ends of the overlap) ([Bisagni et al., 2018](#)). This technique allows to achieve an increase of 50% for failure load performance and a decrease of damage progression for cyclic loads. However, the major drawback associated to this technique is its complexity and the expensive manufacture process it requires.

Stitch insertion

Stitch insertion is another composite through thickness reinforcement technique used which is suitable for improving the mechanical performance of composite bonded joints. This technique can greatly increase the delamination resistance of composites, but the stitching parameters, such as pre-tension and stitch density must be carefully controlled, as they have a significant effect on the peel stresses in the adhesive ([Aymerich et al., 2005](#)). [Tong and Jain \(1996\)](#) have shown that the increase of the pre-tension and the stitch density conduct to a decrease of peel stress present in the adhesive, providing a joint strength improvement when used in SLJs ([Bogdanovich et al., 2011](#); [Reeder and Glaessgen, 2004](#)) and T-joints ([Kim et al., 2017](#); [Bigaud et al., 2018](#)).

The stitch insertion technique has also been shown to be suitable for reinforcing dissimilar SLJs (composite-metal). In this configuration, the stitch thread penetrates through the co-cured adherend to increase the transverse strength of composite, being fixed to holes in the metal adherends. This creates additional load transfer paths that provide a significant contribution to the overall joint strength ([Matsuzaki et al., 2008](#)), although the level of improvement is highly dependent on the pre-tension applied to the stitch.

Conclusions

It is well known that the usage of adhesively bonded joints in many advanced structural applications has increased significantly in the last decades, as this technique provides a very effective method to join composite substrates, enabling the construction of lightweight and durable vehicle frames and components. However, the main drawback associated to the practical implementation

of composite bonded joints is the high likelihood of failing by delamination, that is, a premature failure of the substrates, without extracting the full performance of the materials that compose the joint.

The works published in the literature propose a wide variety of design solutions and techniques to improve the performance of adhesively bonded joints with composite substrates. Among these, the correct selection of the surface treatment and the adhesive to be used are arguably the most important. If a suitable surface treatment is selected, good and durable adhesion between the adhesive and adherend will be attained, ensuring that the load is transferred through the joint. However, it is then important to ensure that this load transfer is not excessively concentrated on a small area of the substrates, which has the potential to generate excessive peel stresses and consequently lead to delamination. The geometrical design of the composite joints is then critical, as it provides several solutions to shape the stress distribution and reduce the peel strength, preventing or delaying the delamination and consequently increasing the joint strength.

A commonly used geometrical feature, which can in some cases be even created naturally, is the use of spew fillet, a simple and effective method to improve the joint strength. Other methods have been shown in practice to be able to improve the joint strength, such as the modification of mechanical properties of the adhesive, the adherend stiffness modification, using a reinforcement of composite with thin layers of metal or polymer in transverse direction, and also the stiffness joint reinforcement that can be achieved by insertion in transverse direction of the joint. While all these techniques allow to obtain a significant improvement in joint strength, they require a more complex and costly manufacture process which, in practice, can be unsuitable for industrial applications. At this stage, and given the increased use of bonded structures of composite construction, an effort is being made to improve the practicality of these methods, with the aim of increasing their practical implementation.

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Dissimilar Joining of PMCs to Metals – Adhesive Bonding

Mariana D Banea, Federal Center of Technological Education of Rio de Janeiro, Rio de Janeiro, Brazil

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Introduction

Adhesively bonded joints are an increasing alternative to mechanical joints in engineering applications and provide many advantages over conventional mechanical fasteners (Budhe *et al.*, 2017). Among these advantages are lower structural weight, lower fabrication cost, and improved damage tolerance (Banea and da Silva, 2009a; Banea *et al.*, 2018a; da Silva *et al.*, 2018b). Design flexibility and joining of dissimilar and/or new advanced materials are also benefits of adhesive joints. Nowadays, most of the industrial structures are made up of different materials (e.g., advanced high strength steel, aluminum, magnesium, fiber-reinforced plastics, sandwich structures etc.), in order to have the best performance (Banea, 2019). The main objective of using material mixes is to allow tailoring the materials in a structure so as to ensure that each part of the structure has the optimum mechanical properties and the minimum weight. PMCs-metal combinations are used in structures to combine the high specific stiffness and strength of metals with the high degree of design freedom of composites. For instance, in automotive industry there is a growing trend to optimize the strength, weight and durability of structures by combining traditional metals with PMCs. Composites are more structurally efficient than metals and do not experience galvanic corrosion, while metals have better damage tolerance and failure predictability than composites and are unaffected by the solvents and temperatures which tend to degrade polymers.

Adhesives can be used to join metals, polymers, ceramics, cork, rubber, and combinations of any of these materials (Banea *et al.*, 2014b). However, these materials need to be joined according to their specific characteristics. Joining dissimilar materials is generally more challenging than joining the same material, mainly because of different coefficients of thermal expansion and potential for galvanic corrosion. In addition, different materials and joint geometries will give rise to distinct stress states in the adhesive layer, and the performance of the adhesive joints will depend on several factors (e.g., surface preparation, the joint geometry and the mechanical properties of adhesive and adherend).

This article gives an overview of recent advances in adhesive bonding joining of PMCs to metals. The main parameters affecting the strength of dissimilar adhesive joints are discussed. Several methods that have been used to predict failure in dissimilar bonded joints are briefly presented.

Parameters That Affect the Strength of Dissimilar Adhesive Joints

The main parameters affecting the strength of adhesively bonded joints are: the surface preparation, joint geometry, material parameters (adhesive and adherend properties), geometrical parameters (adhesive thickness, overlap length, etc.), and environmental conditions (da Silva *et al.*, 2018a; Banea and da Silva, 2009a; Banea *et al.*, 2018a; Budhe *et al.*, 2017). All these parameters must be taken into account during the design of bonded joints with similar and dissimilar adherends.

Surface Preparation of the Adherends

The surfaces play an important role in the bonding process and is, perhaps, the most important process step governing the quality of an adhesive bond joint (Baldan, 2004b; Banea and da Silva, 2009a). Correct surface preparation is essential for good joint strength and maintaining long-term structural integrity of bonded joints. The surface treatment increases the bond strength by altering the substrate surface by increasing surface tension, increasing surface roughness or changing the surface chemistry (Islam *et al.*, 2014; Iqbal *et al.*, 2010; da Silva *et al.*, 2009; Kanerva and Saarela, 2013; Encinas *et al.*, 2014; Sinmazçelik *et al.*, 2011). For PMCs-metal bonded joints, changing surface chemistry of the substrates may result in the formation of a chemical bond between the polymer molecules in the PMCs and the metal oxide layer on the other adherend surface layer (Molitor *et al.*, 2001).

There is a wide range of surface treatments available (Baldan, 2004a; Molitor *et al.*, 2001). However, different industries give different roles to surface preparation. For example, the aerospace industry, with long expected lifetime (several decades), carefully prepares the adherends surface prior to bonding during manufacture and repair to assure the best durability of the bonded structure. On the other hand, the automotive industry with large production scale, prefer to use adhesives that can work with minimal treated surfaces and prefers to overdesign the joints.

Most structural adhesives work as a result of the formation of chemical bonds (mainly covalent, but some ionic and static attractive bonds may also be present) between the adherend surface atoms and the compounds constituting the adhesive. These chemical links are the load transfer mechanism between the adherends. In general, the adhesive bond failures are attributed to poor processes during fabrication, with lack of quality surface preparation being the most significant deficiency (Davis and Bond, 1999). Surface preparation must be tailored to the adherend and will differ for various types of materials. For example, the surface preparation of metals prior to bonding is important due to the oxidization build up that occurs with metals. This is especially important with metals such as aluminum and titanium. Traditional methods of surface treatment such as grit blasting, mechanical

abrasion, and acid etching have been used with good success. In contrast, for plastic materials, sometimes achieving adhesion is quite challenging as plastic surfaces are very smooth, show bad wetting and have low surface energy. Typical composite surface treatments include traditional mechanical abrasion/solvent cleaning techniques for thermoset composites, while thermoplastic composites require surface chemistry and surface topographical changes to ensure strong and durable bond strengths (Molitor *et al.*, 2001).

Mechanical abrasion/solvent cleaning techniques are the most widely applicable surface preparation methods, being suitable for most materials. However, while metal abrasion begins with plastic deformation and work hardening followed by fracture and removal of metal particles, composite matrix resins fracture in a brittle manner during mechanical abrasion with little or no deformation. Mechanical abrasion can remove insoluble contaminants such as mold releases and create a high-energy surface. Grit blasting is another common method used to obtain strong and durable bond strength for metals and thermoset composites. In some cases, to achieve superior durability of the adhesive joint grit blasting in conjunction with a silane is used. For titanium adherends the surface treatments include chromic acid anodisation, sodium hydroxide anodisation methods and laser treatments (Molitor *et al.*, 2001), while for aluminum acetylene and nitrogen plasma treatments can be used before bonding of dissimilar adherends (Rhee and Yang, 2003). For composites, peel ply can be incorporated into the surface of the composite during manufacture and removed prior to bonding (the peel ply must be free of contaminants and release agents).

In summary, surface treatments prior to the application of adhesives are recommended in order to achieve maximum mechanical strength. By increasing surface tension, increasing surface roughness and changing surface chemistry, a more intimate bond can be formed, which allows for increase in strength and durability. The particular surface treatment applied will depend upon the requirements of the bond and the service conditions. The adherend type and geometry and production constraints, as well as cost considerations, may also be relevant factors when selecting a treatment process. In addition, legislative drivers looks to reduce the environmental impact and operator health and safety risks (restrictions have been introduced with respect to the use of certain chemical substances for surface treatments) which promote the need for more environmentally-friendly surface treatments.

Effect of Joint Configuration and Failure Mode

A wide variety of joints are available to the designer of structures. Common joint geometries which have been analyzed in the literature for evaluation of PMCs-metal bonded joints are: Single Lap Joint (SLJ) (Anyfantis and Tsouvalis, 2013; Banea *et al.*, 2018b; Seong *et al.*, 2008), Double Lap Joint (DLJ) (Ban *et al.*, 2008; Wehbe *et al.*, 2019; Kang *et al.*, 2007), Single Strap Joint (SSJ) (Fawzia *et al.*, 2007) and Double Strap Joint (DSJ) (Shin and Lee, 2006; Fawzia *et al.*, 2006; Colombi and Poggi, 2006; Al-Zubaidy *et al.*, 2012; Batuwitage *et al.*, 2017) configurations (see Fig. 1). Peel tests (De Freitas and Sinke, 2014; Arouche *et al.*, 2018) have been also used. For the SLJ test between composite and metal there is a standard available (ASTM D5868-01). However, for the other joint configurations the standardized testing procedures are limited to similar adhesive bonded joints with either composites or metals adherends. Most of the researches adopt the same geometrical configurations described in the standard procedures for similar adhesive joints for testing dissimilar adhesive joints. Nevertheless, the interpretation of the results from the adapted testing methods is not completely clear because of the asymmetric and anisotropic setup (Wehbe *et al.*, 2019).

The strength of a given type of joint depends, for a given type of load, on the stress distribution within the joint. This stress distribution depends on the joint geometry and the mechanical properties of adhesive and adherend (Banea and da Silva, 2009a). In composite-metal adhesive bonded joints, the layered nature of composite adherends and relative weakness in the through-the-thickness direction makes the failure mechanism more complex compared to similar metallic adhesive joints. Due to these uncertainties in joint strength many designers use higher safety margins in their structure resulting in a non-optimum use of materials.

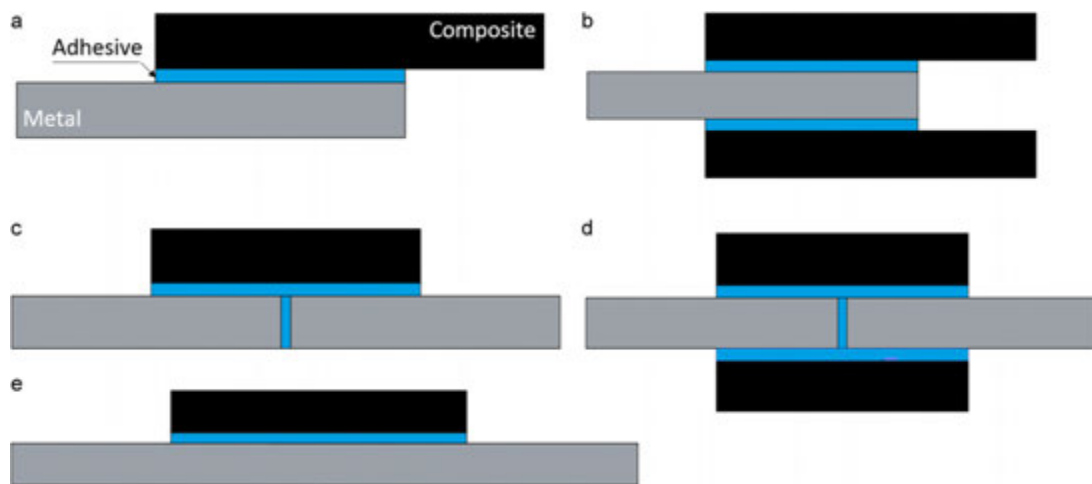


Fig. 1 Examples of configurations of dissimilar adhesively bonded joints: (a) SLJ; (b) DLJ; (c) SSJ, (d) DSJ, (e) reinforced panel (composite patch repair).

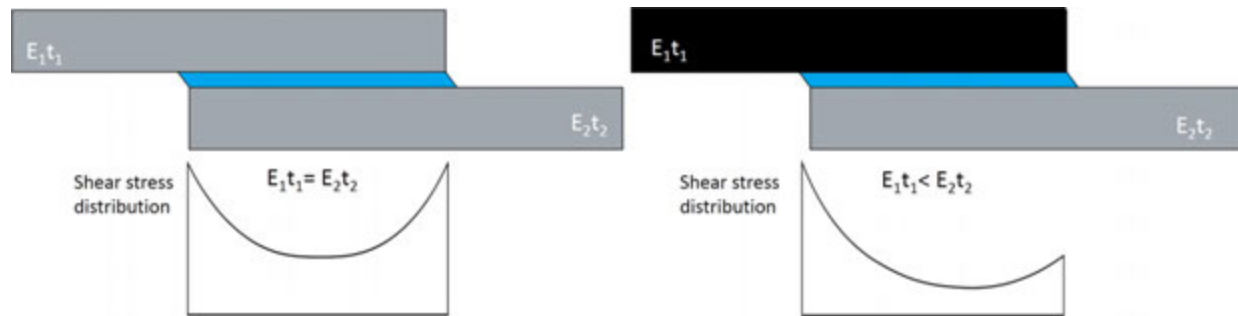


Fig. 2 Effect of the axial stiffness of the adherends on the shear stress distribution.

The single-lap joint is the most common joint used mainly due to its simplicity and efficiency. However, one of the problems associated to this joint is the fact that the stress distribution (shear and peel) is concentrated at the ends of the overlap. It is well known that the adhesive joints with similar adherends are more efficient than the joints with dissimilar adherends (Banea and da Silva, 2009a). The use of dissimilar adherends decreases the joint strength due to a non-uniform stress distribution as can be seen in Fig. 2 (the highest stresses are at the side of the less-stiff adherend, owing to a higher axial strain). In order to mitigate this problem, the joint needs to be designed so that the longitudinal stiffness of the adherends are equal, stiffness balanced joints or the thickness of the two adherends is equal.

Several methods for reducing stress and improving the strength, for similar and dissimilar adhesive joints, have been proposed in the literature. Most involve a geometrical modification of the adhesive, addition of spew fillets and modification of adherend geometry, either by rounding or tapering the adherend edges (da Silva *et al.*, 2018b). For example, Belingardi *et al.* (2002) found that the stress concentration (shear and peeling components) are lowered with the decrease of the spew fillet angle, although most of the advantage is obtained within the angle 45° solution. It is concluded that shaping the spew fillet and increasing its size can provide smoother transition in joint geometry, significantly reducing the stress concentration. Rispler *et al.* (2000) obtained a 66% reduction of peel stress by fillet shape optimization in CFRP/titanium DLJs. Many of these methods, however, increase the design and manufacturing complexity.

In practice, adhesive joints are subjected to thermal as well as structural loads. The mismatches in the thermal and mechanical properties of the adhesive and adherends cause deformations and stresses induced by thermal fields in all members of the adhesive joint, especially in the adhesive layer (Sen, 2016). Thermal loads are especially important when bonding dissimilar materials (with different coefficients of thermal expansion), as large differences in thermal expansion characteristics between adherends can cause severe problems (Rastogi *et al.*, 1998). For example, for composite-aluminum bonded joints, the difference in thermal expansion between the adherends is relatively large, giving considerably higher thermal stresses. In addition, CFRP has a particularly low thermal expansion, so that when bonded to metals these materials tend to produce higher thermal stresses than when are bonded to other materials. If a ductile adhesive is used, the adherends can contract freely and no significant stresses arise in the joint. However, when the adhesive is stiff (e.g., epoxy), the composite adherend will be under a compressive load and the metal will be subjected to a tension load. This leads to bending of the joint, and the stress field in the joint will be composed of the uniform axial load plus a new bending component (Marques *et al.*, 2014). Rastogi *et al.* (1998) studied three-dimensional (3D) thermal stress distributions in composite-aluminum, symmetric, double lap joints subjected to uniform temperature loads. They found that the joint corners are critical regions for debonding initiation. Owens and Lee-Sullivan (2000b) studied stiffness loss due to crack growth in composite-to-aluminum joints. They tested single lap joints at room temperature and at -40°C at quasi-static conditions and found that the joint stiffness is more affected by the response of the adherends to the test temperature than by the modulus of the thin adhesive layer.

To summarize, in order to minimize the effect of thermal loads, the joint geometry should be optimized to allow the materials some freedom to expand without transferring stresses, by leaving larger gaps between the substrates or by modifying the adhesive thickness, for example. The correct selection of the adhesive is also an important method to minimize the thermal stresses. By selecting ductile adhesives, able to handle the different levels of thermal expansion, the substrates can be joined without significant transfer of stresses.

There are several possible failure modes in a PMCs-metal bonded joint subjected to a tensile force (see Fig. 3): metal and adhesive interface debonding, cohesive failure (adhesive layer failure), composite and adhesive interface debonding, composite delamination (separation of some fibers from the resin matrix), composite rupture and metal yielding. The failure modes of adhesive joints depend on the types of the adherend and adhesive. Adherend yielding might occur in structures where metals such as aluminum or mild steel are used. On the other hand, unlike isotropic adherends, PMC adherends have relatively low transverse strength and shear stiffness compared to their in-plane material properties and it is, therefore, important to understand the mechanisms of failure involved for each specific material. Seong *et al.* (2008) investigated the influences of bonding pressure, overlap length, adherend thickness, and type of adherend material on the failure mode of composite-aluminum SLJs. Failure modes of these joints were complicated with a combination of delamination failure and interface failure. However, delamination was the major failure mode. Zhao and Zhang

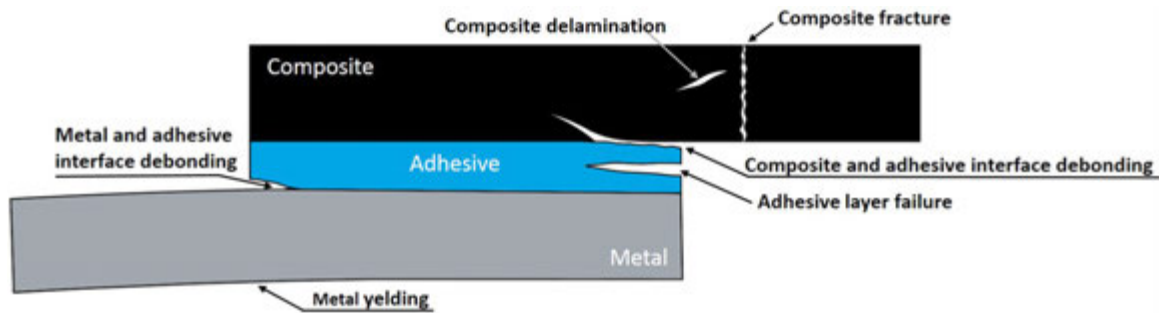


Fig. 3 Failure modes in a composite-metal bonded joint.

(2007) states that the debonding failure is the most common failure in FRP-steel joints in case of a proper design for steel cross section.

Effect of Mechanical Properties of Adhesive and Adherend

The adhesive and adherend material influence the adhesively bonded joints performance and its effect should be taken into consideration in the design of similar and dissimilar adhesive joints.

There are many different types of adhesives available, each with their own advantages and disadvantages: epoxies (having high strength and temperature resistance), cyanoacrylates (fast bonding capability to plastic and rubber but poor resistance to moisture and temperature), anaerobics (suitable for bonding cylindrical shapes), acrylics (versatile adhesives with capabilities of fast curing and tolerate dirtier and less prepared surfaces), polyurethanes (good flexibility at low temperatures and resistant to fatigue) and high-temperature adhesives (phenolics, polyimides and bismaleimides). The selection of a suitable adhesive is not an easy task, as there is no universal adhesive that will fulfill every application and the selection of the proper adhesive is often complicated by the wide variety of available options. Adhesive selection includes several factors such as: type and nature of substrates to be bonded, cure and adhesive application method and the expected environments and stresses that the joint will face in service. Also, the cost of the adhesive may sometimes be an important factor of adhesive selection in a particular production situation (Banea *et al.*, 2018a). The first criteria for choosing an adhesive is that it can create a proper bond between the materials chosen for the structure. Datasheets from adhesive manufacturers provide basic information about what type of materials can be bonded. Some adhesive suppliers provide software tools to help the engineer or designer to select the adhesive, although for critical components expert assistance is still needed.

For joining dissimilar materials, the selection of a proper adhesive is more critical because of the thermal deformation and stresses caused by the difference in thermal expansion coefficients of the materials, which may result in the premature failure of the joints and a reduction in joint strength. To reduce the thermal stresses, elastomeric and ductile adhesives can be used. It was shown that ductile adhesives increase the resistance to crack growth, thus increasing the joints' strength of composite-to-aluminum joints, (Owens and Lee-Sullivan, 2000a,b). However, using low modulus ductile adhesives can lower the joint rigidity. Another solution is to use co-curing during joining (using the resins used to produce the polymer composite as an adhesive which bonds the composite and metal parts), which ensures that curing and joining take place at the same time. This process doesn't need an additional curing process and in this way the labor consumption is reduced. However, co-curing did not promote a significant increase in joints strength when compared to conventional (secondary) adhesive joints.

The adherend material is another important parameter that influence the adhesively bonded joints performance. Most of the published work examines the influence of adherend stiffness on the joint strength and, in general, increasing the adherend stiffness improved the joint's strength (Kafkalidis and Thouless, 2002; Karachalios *et al.*, 2013a,b; da Silva *et al.*, 2009; Reis *et al.*, 2011; Rispler *et al.*, 2000). The adherends can have different flexural stiffness due to either geometric and/or material considerations. It was shown that for low strength adherends, an increase in thickness is beneficial because the adherend becomes stronger and less likely to deform plastically, while for high strength adherends, a higher thickness can decrease the joint strength due to an increase of the bending moment (Banea, 2019).

The effect of different material combinations on strength of dissimilar bonded joints was extensively investigated in the literature (Anyfantis and Tsouvalis, 2013; Di Franco *et al.*, 2013; Hasheminia *et al.*, 2019; Campilho *et al.*, 2018; Ribeiro *et al.*, 2016). Owens and Lee-Sullivan (2000a,b) studied the behavior of polymer composite-aluminum joints, and developed an analytical model to predict the joint stiffness and respective loss with the crack growth. They found that flexible adhesives increase the resistance to crack growth, thus increasing the joints' strength. Rudawska (2010) performed an experimental and numerical study on joints between different adherend materials (titanium, aluminum alloys and aramid-epoxy composites). The joints strength varied depending on the chosen adherend combinations, with the best results for the aluminum-aluminum joints. Gultekin *et al.* (2014) showed that in composite-aluminum dissimilar joints the stacking sequence and thickness of the composite in adhesively bonded substantially influences the load-carrying capacity of the joint. More recently, Banea *et al.* (2018b) investigated experimentally and numerically dissimilar adhesive joints by using various combination of adherends

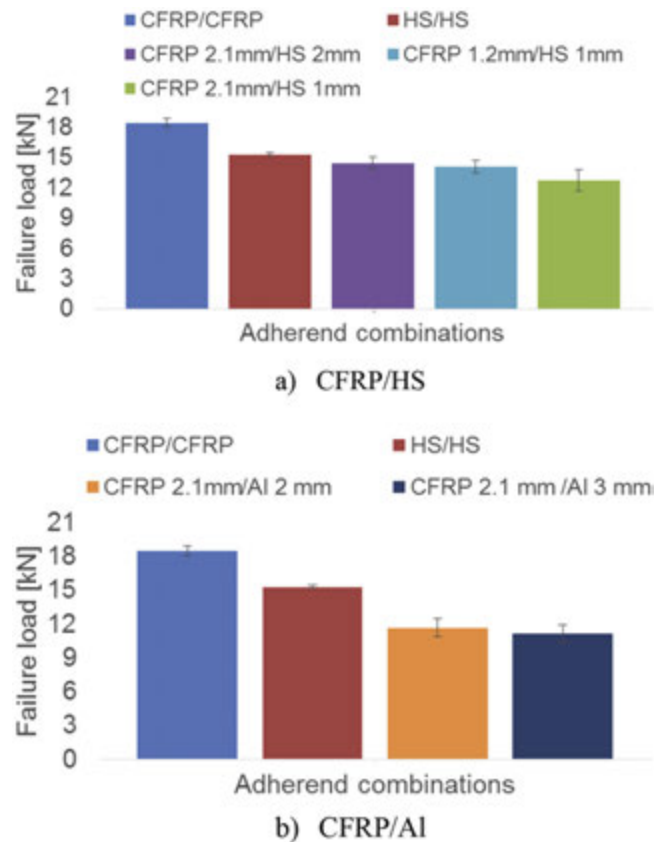


Fig. 4 The effect of material combination on the failure load of multi-material SLJs. Reproduced with permission from Banea, M.D., Rosioara, M., Carbas, R.J.C., da Silva, L.F.M., 2018b. Multi-material adhesive joints for automotive industry. *Composites Part B: Engineering* 151, 71–77.

(high strength steel (HS), aluminum (Al) and carbon fiber reinforced plastics (CFRP)) bonded with a modern tough structural adhesive used in the automotive industry. The experimental work was accompanied by a numerical analysis by Finite Element Method (FEM), based on Cohesive Zone Models (CZM). A bilinear cohesive damage model was used to obtain the numerical predictions as a function of material combination. It was found that geometrical dissimilar balanced joints ($t_{s1} = t_{s2}$) promote a higher strength than the stiffness dissimilar balanced joints ($E_1 t_{s1} = E_2 t_{s2}$). The most efficient joint was the joint with similar CFRP adherends, becoming less so for dissimilar adherends, particularly for the case of unbalanced geometrical joints (i.e. CFRP (2.1 mm)/HS (1 mm)) as can be seen in [Fig. 4](#) (Banea *et al.*, 2018b).

Effect of Adhesive Thickness and Overlap Length

The adhesive thickness is also an important parameter that influences the adhesively bonded joints performance (Banea *et al.*, 2015). For SLJs, it was shown that the joint's strength decreases as the adhesive thickness increases, with the exception of SLJs bonded with elastomeric adhesives (Banea and da Silva, 2010b, 2009b). The highest strength is obtained for adhesive thicknesses of the order of 0.1–0.5 mm (da Silva *et al.*, 2006; Gleich *et al.*, 2001). Moreover, this observation is not generally applicable as there are other factors involved, such as the type of loading, the adherend behavior (elastic or plastic), and the type of adhesive (ductile or brittle), which can modify the behavior of joints as their thickness is varied (Banea *et al.*, 2015).

The overlap length is another parameter that can affect the joint strength (Banea and da Silva, 2010a). While increasing the joint width increases the strength proportionally, the effect of the overlap length on the adhesive joint strength depends on the type of adhesive (i.e., ductile or brittle) and also on the type of adherend (Banea *et al.*, 2017). For instance, for bonded joints with elastic adherends and ductile adhesives, the strength is proportional to the overlap length. This occurs because ductile adhesives deform plastically as the load increases, and make use of the whole overlap. In this case the global yielding failure criterion is suitable. For joints with elastic adherends and brittle adhesives, the joint strength is not proportional to the overlap and a limit strength is attained. In this case the adhesive does not accommodate peak stresses at the ends of the overlap and failure is ruled by these peaks. On the other hand, for composite adherends the overlap effect on the strength of the joints is mainly dictated by the composite transverse strength (Neto *et al.*, 2012). One of the issue in bonding composite adherends is the delamination, due to the fact that composites are made of stacked layers. This is particularly true if the composite adherends are made of layers with

different fiber orientations. It is well known, that in the fiber direction, unidirectional composites can be very strong and stiff, whereas the transverse and shear properties are much lower. Thus, in order to avoid premature failure of the composite adherends it is recommended to have the outer layers of the adherend with a direction parallel to the loading direction to avoid intralaminar failure of these layers. Seong *et al.* (2008) found that for composite-aluminum bonded joints with FM73 adhesive, the failure load is not linearly proportional to the overlap length. When the overlap length was larger than 25 mm, or overlap length-to-width ratio was larger than 1, the failure load did not increase substantially. They concluded that high efficiency might not be obtained when the ratio of overlap length to the width of single-lap bonded joints is much larger than 1.

Environmental Conditions

Adhesively bonded joints may be exposed to various environmental conditions during their service life. It was shown in the literature that the performance of adhesive systems can be considerably deteriorated when exposed to harsh environments. The environmental factors must be considered as critical in determining the long-term durability of adhesively bonded joints and need to be carefully identified and related to the type of service the material will see.

The main environmental factors are the temperature and humidity. The prolonged exposure or even short term exposure to elevated temperatures will often produce irreversible chemical and physical changes within adhesives. As the temperature increases, the bond strength decreases (Banea *et al.*, 2012, 2014a; Marques *et al.*, 2014). Also, the moisture absorbed in a polymeric material can lead to a wide range of effects, both reversible and irreversible, including plasticization, swelling, and degradation. At temperatures below the glass transition temperature (T_g), polymer property reduction is reversible upon dehydration, whereas above T_g , the properties are permanently degraded (Viana *et al.*, 2017).

In general, moisture is known to increase the ductility and reduce the elastic modulus and strength of resins/adhesives. However, the presence of moisture in adhesive joints may not only weaken the physical and chemical properties of the adhesive itself but also the interface between the adhesive and the substrate. In fact, it was shown that the degradation of the interface is often significantly larger than that of the adhesive (Loh *et al.*, 2002).

For PMCs-metal bonded structures, the influence of environmental aspects has specific relevance, as composites and metals have different reactions to the same environmental conditions. Temperature variations and cycles can affect the performance of composite and composite-metal bonded structures. It is generally accepted that the resin matrix, adhesive and fiber/matrix interface are the components in adhesively bonded PMC-metal joints that are most susceptible to thermal effects. However, due to a wide range of produced PMCs (with a large number of possible fiber/matrix combinations) and structural adhesives, it is extremely difficult to make any generalization when it comes to the effect of temperature on PMCs-metal bonded joints. For instance, Zhou *et al.* (2019) investigated the effects of mechanical and geometrical parameters of a CFRP-to-steel bonded joints at elevated temperatures on bond strength. They found that for bonded joints with sufficiently long bond length, the ultimate load depends only on the fracture energy of the final temperature, and the maximum load of the bonded joints depends on the ratio between the loading and heating rates. Qin *et al.* (2018) have also found that high and low temperatures can have direct influence on the strength failure mode of dissimilar composite-aluminum joints. Avendaño *et al.* (2016) investigated the joint strength of SLJs made of dissimilar lightweight adherends (unidirectional carbon fiber reinforced polymer and an environmentally friendly biopolymer) under quasi-static and impact loading as a function of temperature. They found a significant decrease on joint strength at high temperature. More recently, Machado *et al.* (2019) investigated the behavior of dissimilar adhesive joints, using composite and aluminum substrates, under quasi-static and impact loads at different testing temperatures (−30 to 80°C), following the requirements for the automotive industry. They found that dissimilar adhesive joints, if used in conjunction with modern crash resistant adhesives, can effectively be used for the construction of automotive structures, without significant sacrifices in joint performance, with good energy absorption capabilities under impact.

Regarding the effect of humidity, it was shown that, for composite joints exposed to humid environments, the mechanisms of degradation are different compared to metal joints. Unlike metals, the work of adhesion for composite to epoxy joints remains positive in the presence of water, thus decreases the likelihood of interfacial failure on ageing. Also, the composite adherend will absorb water, which can affect the kinetics of water absorption into the adhesive. In addition, the interface between fibers and matrix may be weakened in the presence of moisture (Budhe *et al.*, 2018). On the other hand, when metallic adherends are used, moisture might degrade the adhesive-adherend interface. In order to eliminate or diminish this damage, a suitable surface pre-treatment should be used (Critchlow and Brewis, 1996).

Although, the separate effect of temperature and moisture is relatively well understood, the combined effect of temperature and moisture is not yet fully understood. The combined effect of moisture and temperature (hydrothermal ageing), is known to be among the most severe exposure conditions. Moisture is responsible for lowering the glass transition (T_g) of the adhesives and this has an influence on their behavior, especially at high temperatures. At lower temperatures, as the adhesive is already well below T_g , its influence is not as significant (Viana *et al.*, 2016). Heshmati *et al.* (2017) investigated the behavior of CFRP/steel and GFRP/steel double-lap shear joints, subjected to a number of environmental conditions for up to three years. They found that immersion at 45°C resulted in higher strength reductions of the joints strength (see Fig. 5). The strength and stiffness of the joints made of GFRP material underwent significant reductions, while the CFRP/steel joints were affected to a considerably smaller degree. In addition, the use of polyester instead of epoxy resin as the matrix of FRPs was found to result in the higher degradation of joints exposed to hot/wet conditions, in particular.

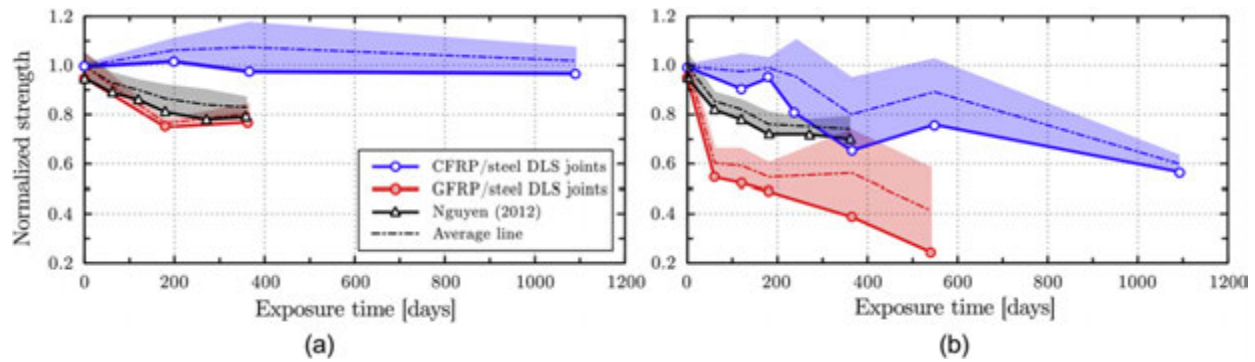


Fig. 5 Effect of immersion conditions at different temperatures on the strength of DLS joints: (a) 20°C, (b) elevated service temperatures (45–50°C). Reproduced with permission from Heshmati, M., Haghani, R., Al-Emrani, M., 2017. Durability of bonded FRP-to-steel joints: Effects of moisture, de-icing salt solution, temperature and FRP type. *Composites Part B: Engineering* 119, 153–167.

Although, various studies were employed on the effects of various environments on the adhesive properties (Viana *et al.*, 2017), it is still necessary to address the performance of specific adherend-adhesive combination. Adhesive joints are systems comprised of adherends, adhesives, and inter-phase regions and the performance of each of these components may strongly affect the performance of the bonded joint. Therefore, general knowledge of the behavior of adhesives exposed to various environments must be supplemented by knowledge of the behavior of specific bonded materials. The influence of environmental aspects has specific relevance for multi-material structures, as different materials have different reactions to the same environmental conditions and can significantly alter the behavior of the bonded structure as a whole. This requires a renewed investigation of ageing effects, as well as a survey on the influence of environmental conditions like temperature, humidity etc., to which these materials may be more sensitive.

Analysis of Dissimilar Adhesively Bonded Joints

In order to design structural joints in engineering structures, it is necessary to be able to analyze them. This means to determine stresses and strains under a given loading, and to predict the probable points of failure. There are two basic mathematical approaches for the analyzes of adhesively bonded joints: closed-form analysis (analytical methods) and numerical methods (i.e., finite element analyses). The analysis of adhesive joints started almost 90 years ago with the closed-form model of Volkersen (1938) that considered fully elastic materials and adhesive deformation only in shear. However, the analytical formulation of adhesive joints becomes more complex if the adhesive deforms plastically, if composite adherends are used, or if dissimilar adherend materials (e.g., composites with plastic metals) are used. Numerical methods (e.g., finite element method) are commonly used to perform non-linear stress analyses and predict the joints strength, traditionally by using stress/strain or fracture mechanics criteria. These techniques showed acceptable results, but have some limitations: stress/strain methods are mesh dependent, while fracture criteria are restricted to Linear Elastic Fracture Mechanics (LEFM) and rely on the existence of an initial crack. One method that surpasses these limitations is the CZMs. This method has received considerable attention in the last years and has been employed for a wide variety of problems and materials including metals, ceramics, polymers and composites. A CZM models the fracture process extending the concept of continuum mechanics by including a zone of discontinuity modeled by cohesive zones, thus using both strength and energy parameters to characterize the debonding process along the crack path, allowing the approach to be of much more general utility than conventional fracture mechanics. CZMs may be used for adhesive debonding or for composite delamination, when composite adherends are used. However, the cohesive models present a limitation, as it is necessary to know in advance the critical zones where damage is prone to occur and place the cohesive elements in accordance. Continuum damage models constitute a valuable alternative when crack path is not known in advance. Also, continuum damage models acquire special relevancy when adhesive thickness has to be considered (Banea, 2017).

Several authors (Anyfantis and Tsouvalis, 2013; Rispler *et al.*, 2000; Fernandes *et al.*, 2015) conducted a series of tests and CZM simulations of adhesive joints between similar and dissimilar adherend materials. Anyfantis and Tsouvalis (2013) modeled the joints between CFRP composites and steel bonded with a ductile adhesive layer. The elasto-plastic loading and fracture response were modeled by a recently developed mixed-mode CZM law. Campilho *et al.* (2018) and Ribeiro *et al.* (2016) used CZM modeling to study the joints' strength and stress analysis of CFRP-aluminum adhesive joints bonded with a brittle and a ductile adhesive. They concluded that the joints' strength and failure modes were highly dependent on the adhesive. Banea *et al.* (2017) investigated experimentally and numerically the effect of material on the fracture behavior of adhesive joints bonded with modern tough structural adhesives. SLJ tests with various adherend materials (high strength steel, low strength steel and composite) were performed. The experimental work is accompanied by a numerical analysis by FEM, based on CZM. A bilinear cohesive damage model was used to obtain the numerical predictions as a function of material for each adhesive. The CZM returned satisfactory results for short overlaps, but with large overlaps these models did not work with the same precision. It was concluded that fracture parameters as a function of material are needed.

Hua *et al.* (2012) proposed a strain-based failure model to deal with progressive cohesive failure in composite-aluminum SLJs for a range of environmental degradations. Their model can predict not only the failure loads of the joints but also the damage initiation and propagation in the degraded adhesives. The elastic-plastic response of the adhesive and the substrates, both obtained from the bulk tensile tests, were incorporated in the coupled mechanical-diffusion analyses. The main limitation of the model is the mesh dependence of the material failure parameters. They proposed a displacement based continuum damage model in order to overcome this mesh dependence, validated by undertaking progressive damage modeling on SLJs and found good correlation between predicted and experimental results, demonstrating that the continuum damage model is an efficient and reliable method to model environmental degradation in ductile adhesive bonded joints, where failure is predominantly within the adhesive layer (Hua *et al.*, 2008).

Moreover, in general, complex material models are required to accurately analyze the structural failure behavior of bonded joints subject to service conditions (e.g., temperature and humidity) (Viana *et al.*, 2017). Currently, there is no temperature or/and moisture dependent CZM available in any commercial finite element software. As a result, the simulation should be performed with a set of parameters adjusted to the temperature/moisture dependent fracture problem. These aspects are under intense research nowadays and the development of reliable design and predictive methodologies it is believed to result in more efficient use of materials and adhesives in industry.

Summary

Nowadays, new products consist more and more of a combination of advanced materials, which provides an opportunity to develop structures capable to operate under more exigent requests demanded by industry such as: increased strength-to-weight ratio, multifunctionality and low carbon emissions. These materials need to be joined according to their specific characteristics. Therefore, combining multiple materials to create lightweight yet durable structures packed with distinctive design features can be a challenge.

Dissimilar material combinations for multi-material structures will often be joined using adhesive bonding technology. Successful adhesive bonding of PMCs-metal structures relies on good surface treatment of both the composite and metal and suitable selection of the adhesive that can compensate for the differences in thermal expansion of the substrates and has good mechanical characteristics. The adhesive must be able to form a good bond with both types of substrate, have good mechanical characteristics and be able to transfer the load between the dissimilar substrates effectively.

The failure modes of adhesive joints depend on the types of the adherend and adhesive. Adherend yielding might occur in structures where metals such as aluminum or mild steel are used. On the other hand, unlike isotropic adherends, PMC adherends have relatively low transverse strength and shear stiffness compared to their in-plane material properties and it is, therefore, important to understand the mechanisms of failure involved for each specific material. A good failure criterion should predict both failure mode as well as failure load. However, the accurate prediction of the joint strength has been limited by the lack of a suitable, universally applicable failure criterion and insufficient accuracy in calculating the stresses and strains in the adhesive layer. In addition, complex material models (and consequently material data) are necessary to accurately analyze the failure behavior of dissimilar bonded joints subject to service conditions.

The influence of environmental conditions has specific relevance for dissimilar bonded joints, as different materials have different reactions to the same environmental conditions and can significantly alter the behavior of the bonded structure as a whole. Long-term performance and uncertainty relating to environmental durability still remain a barrier to the wide application of composite and PMC-metal bonded joints in structural applications. This topic is under intense research at the moment and the development of reliable design and predictive methodologies can be expected to result in more efficient use of materials and adhesives.

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Mechanical Properties and Non-Destructive Evaluations of Joints Based on Polymer Composites

Pietro Russo, Institute for Polymers, Composites and Biomaterials, National Research Council, Pozzuoli, Italy

Ilaria Papa and Valentina Lopresto, Department of Chemical, Materials and Production Engineering, University of Naples Federico II, Naples, Italy

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Glossary

Adhesive Joint A joint connected by an adhesive (glue-like) material.

Butt Joint A joining method in which two components are simply placed end-to-end together without any shaping.

Fastener Device (eg rivet, bolt, screw) that mechanically joins two or more components.

Friction Welding A solid welding process that produces coalescence of material by the heat obtained from a mechanically induced sliding motion between rubbing surfaces. The work parts are held together under pressure.

Net section Net or available area of a cross section of a beam after deducting the holes drilled for rivets, bolts and so on.

Residual Stress Stress remaining in a structure or member, as a result of thermal and/or mechanical treatment.

Tensile Strength The maximum stress a material subjected to a stretching load can withstand without tearing.

Weldability The capacity of a material to be welded under the fabrication conditions imposed into a specific, suitably designed structure and to perform satisfactorily in the intended service.

Preface

Composite materials are nowadays applied mainly in several fields from aerospace to biomedical and sports due to the high mechanical performances together with their lightness. They mostly consist of three different phases: reinforcement, matrix and interfaces. The former is usually in the form of particles characterized by a specific aspect ratio or fibers of natural, mineral or synthetic origin, which can be short, long or even woven into fabrics with various architecture. The matrices, on the other hand, are usually polymeric but also of the ceramic or metal type, while the interfaces play a fundamental role in the transmission of the stresses, at which composites are subjected mainly in conditions of use, between the phases mentioned above. These features lead to the no homogeneity and the anisotropy of these materials: aspects that simultaneously constitute both their strengths and their weaknesses.

Since composite material means innovative material made by different constituent, several different kinds of composites are possible to fabricate depending on the reinforcing and the matrix. In the present article special attention will be paid on fiber-reinforced polymers (FRP) being the most used ones due to their relatively low weight together with their very high performances.

The impregnation in a polymeric matrix of reinforcement with different orientation and architecture allows obtaining a material with mechanical properties depending on the direction and the disposition in plane and along the thickness of the fibers. It permits to satisfy at the best the specific requests in terms of loading direction and to create materials “ad hoc” for specific applications. On the other side, the anisotropy represents the most crucial aspect from the damage point of view, leading to different failure modes and several different interactions between them under loading conditions, not always predictable and not always detectable.

In this complex scenario, the design of the joint in FRP is a very critical issue. The joint is a source of stress concentration, and improper design of the same can enhance this drawback causing its premature failure.

For FRP composites, three main types of joints can be distinguished: *Adhesive Joints* (permanent), where an adhesive joins two substrates, *Mechanical Joints* (temporary), where rivets, bolts and screws are used and *Welded Joints* (permanent) achieved by various method as ultrasonic vibration, electromagnetic induction and thermal techniques, commonly used to assembly thermoplastic and metal composite components. It is also possible to consider a combination of these methods to get additional benefits (*hybrid joints*).

The following three paragraphs give a brief description of these joint methodologies, highlighting their advantages, limitations and the main parameters that influence their quality as well as their typical failure modes.

Adhesive Joints

Joints realised between two substrates called adherends by using an adhesive that is a polymer generally selected depending on the basis of the nature and other chemical and physical aspects of the materials to be combined, applications, service environment and costs. Single lap, tapered lap, scarf, butt, strap and double strap, tapered double strap, double lap and stepped lap fall in this type of joints.

There are several manufacturing bonding processes, namely (*Ata et al., 2019*) co-curing without adhesive, co-curing with adhesive, secondary bonding, and co-bonding on the shear strength of composite single-lap joints (*Vallée et al., 2017*) to join composite substrates (*Fig. 1*).

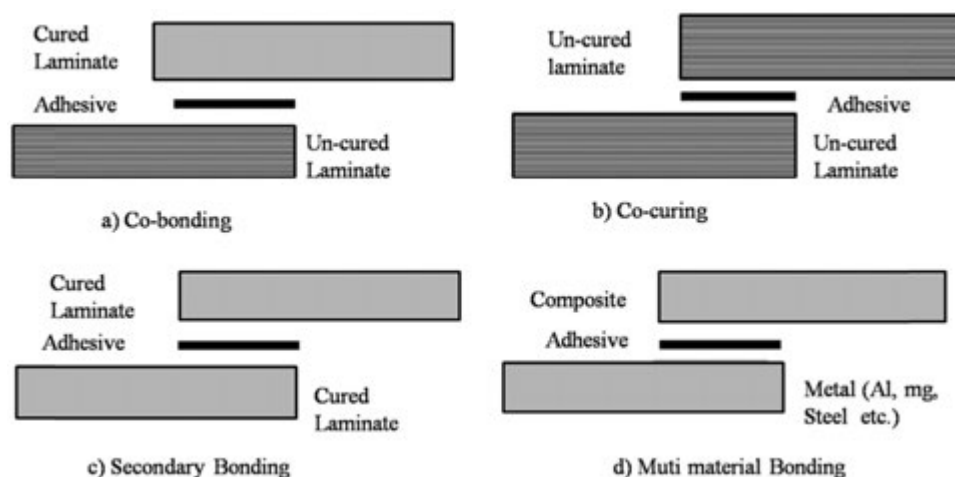


Fig. 1 Schematic of the common manufacturing bonding processes between composite components. Reproduced from Budhe, S., Banea, M.D., de Barros, S., da Silva, L.F.M., 2017. International Journal of Adhesion and Adhesives 72 (October), 30–42.

Secondary bonded joints have higher strength than co-bonded and adhesively co-cured joints and give rise to similar strength compared with the non-adhesive co-cured case. Co-cured joints without an adhesive show the highest strength (Ashcroft *et al.*, 2006) and satisfactory performance both at high temperatures and in wet conditions while co-bonded joints exhibit maximum strength at low temperatures. In the co-bonding process, an adherent is polymerized with the adhesive, while both parts are simultaneously polymerized in a co-curing process. When the adhesive layer is polymerized between two pre-polymerized substrates, a so-called “secondary bonding” is obtained.

In industrial applications, a wide variety of joint configurations is used (Mahi *et al.*, 2014). Stress concentrations influence the strength of the joint at the end of the overlap. Reminding that the geometry of the joints can reduce the stress concentration (Neto *et al.*, 2012; Sinmazçelik *et al.*, 2011; Borsellino *et al.*, 2009; da Silva *et al.*, 2009; Kanerva and Saarela, 2013), various techniques have been proposed to reduce peel and interfacial stresses (eg spew fillet, adhesive thickness, mixed adhesive, conical plate with different ends, different thickness, tapered width, length and thickness, etc) (Islam *et al.*, 2014; Iqbal *et al.*, 2010; Encinas *et al.*, 2014; Kreling *et al.*, 2013; Fischer *et al.*, 2012).

Overall, all the geometrical parameters (eg adherend thickness, width, adhesive thickness, tapered length, tapered thickness, stacking sequence, ply angle, fillet and so on) influence the bonded joints performance, so optimization of them is necessary to maximize their mechanical strength. At this regard, beneficial effect of an internal taper endplate and bi-adhesive in lowering the stress concentration at the end of the overlap have been demonstrated (Mahi *et al.*, 2014; Neto *et al.*, 2012). However, manufacturing is a crucial point. For example, it has been noted that applying the taper on a small part rather than along the entire adhesive layer is more efficient (Fig. 2).

The chemical and physical surface properties can be changed by applying a thin coating. At this regard, different treatments are available and a proper selection among them is crucial (Sinmazçelik *et al.*, 2011; Borsellino *et al.*, 2009; da Silva *et al.*, 2009; Kanerva and Saarela, 2013) since the surface preparation strongly influences the quality of the bonded joint. This step should remove all the contaminants (lubricants, dust, loose corrosion layers, micro-organisms) from the surfaces, ensure their good wettability, functional activation of material surfaces being bonded, etc (Islam *et al.*, 2014; Iqbal *et al.*, 2010; Encinas *et al.*, 2014; Kreling *et al.*, 2013; Fischer *et al.*, 2012). Furthermore, a proper surface treatment of the substrate before adhesive bonding plays an essential role in the durability of the bonded joints (Pereira *et al.*, 2009; da Silva *et al.*, 2010b; de Barros *et al.*, 2017; Dawood and Rizkalla, 2010; Rechner *et al.*, 2010; Brack and Rider, 2014; Marzi *et al.*, 2011; Boutar *et al.*, 2016). A careful selection of the surface preparation method concerning the substrate material is needed, as some adhesive joining methods may degrade their characteristics. For example, given the significant dependency on surface roughness (da Silva *et al.*, 2010b), it is essential to focus on this issue to improve the fatigue life of adhesive joints, as most of the structures are subjected to dynamic loading.

Another relevant geometric parameter is the thickness of the adhesive layer. At this regard, research efforts are available in the literature about the adhesive-bonded joint fracture under mode-I using double-cantilever beam (DCB) joints (de Barros *et al.*, 2017; Rechner *et al.*, 2010; Brack and Rider, 2014; Marzi *et al.*, 2011; Boutar *et al.*, 2016; Costa-Mattos *et al.*, 2010; Dawood and Rizkalla, 2010; Azari *et al.*, 2011; Carlberger and Stigh, 2010; Ji *et al.*, 2013; Cooper *et al.*, 2012), tapered double-cantilever beam (TDCB) joints (de Barros *et al.*, 2017; Naito *et al.*, 2012) and butt joints (da Silva *et al.*, 2010a; Tang *et al.*, 2013). Mode II has also been studied using the end-notched flexure joints (Costa-Mattos *et al.*, 2010; Azari *et al.*, 2011; Xu and Wei, 2013), while mixed mode loading was assessed through single lap joints (SLJ) (Tang *et al.*, 2013; Xu and Wei, 2013; Castagnetti *et al.*, 2011; Xu and Wei, 2012) for which, in general, the strength decreases as the adhesive thickness increases (Banea *et al.*, 2014b). This effect can be ascribed to more defects like voids, microcracks and higher interface stresses, contained in a thicker bond line (Costa-Mattos *et al.*, 2010; Tang *et al.*, 2013; Barus *et al.*, 2018; Arenas *et al.*, 2010).

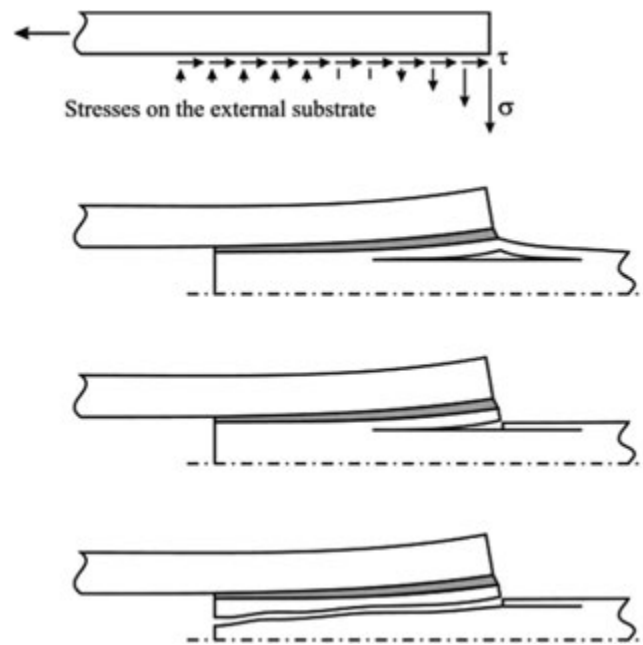


Fig. 2 Failure in adhesive double-lap joints due to transverse (through the thickness) stresses of the composite substrates. Reproduced from Banea, M.D., da Silva, L.F.M., 2009b. *Proceedings of the Institution of Mechanical Engineers, Part L: Journal of Materials: Design and Applications* 223 (1), 1–18.

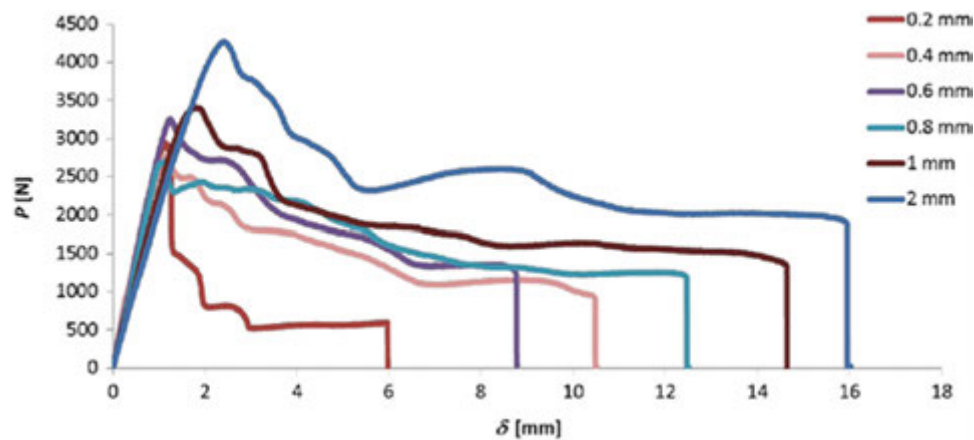


Fig. 3 Representative experimental P–d curves as a function of adhesive thickness. Reproduced from Banea, M.D., da Silva, L.F.M., Campilho, R. D.S.G., 2014b. *Journal of Adhesion* 91 (5), 331–346.

However, there is no any generalized trend between strength and adhesive thickness due to various parameters involved like the type of loading (mode I, mode II, or mixed), the adherend behavior (elastic or plastic), type of adhesive (ductile or brittle), geometry of joints etc (Ji *et al.*, 2013; Tang *et al.*, 2013; Suwanpakpraek *et al.*, 2019; Liao *et al.*, 2013). and the identification of specific correlations can be quite complex (Fig. 3).

The failure strength is higher in thicker adherend joints and lowers in specimens with larger overlap lengths (Neto *et al.*, 2012). It was shown (Li *et al.*, 2015) that a 0.5 mm adhesive thickness is optimum by using 2 mm adherend thickness, Henkel Loctite 330 as adhesive, and an overlap length of 25 mm on tensile lap-shear strength of single overlap joints. Moreover, the shear strength increases as the thickness of the adhesive are far below 0.4 mm.

Generally, in order to optimize the adhesive thickness it is important to consider properties of both the adhesive (Reis *et al.*, 2015; Abdel Wahab, 2012) and the adherend material (Meneghetti *et al.*, 2010), the stacking sequence and fiber orientation of the different plies (Hazimeh *et al.*, 2015; Ozel *et al.*, 2014; Jen and Ko, 2010; Banea *et al.*, 2015), moisture content, the presence of any reinforcing fillers added to improve specific mechanical parameters and also the type of loading (Banea *et al.*, 2014d). In practice,

this optimization phase is further complicated by the application of mixed load conditions: an aspect that represents a significant gap in the literature.

Some Parameters Affecting the Performance of Adhesive Joints

The adhesive

The selection of adhesive materials for a specific application is not an easy task as it depends on many factors, i.e. adherend type to be bonded, curing temperature, expected environmental condition during service, type of load, cost etc. The reuse, recycling and recovery of bonded parts are nowadays the major concerns: the adhesive bonding should quickly disband without damaging the structure.

For example, the functional additive approach by chemical foaming agents and thermally expandable particles (TEPs), as well as the pressure sensitive adhesives, can be introduced quickly into existing adhesive products (Sun *et al.*, 2013; Banea *et al.*, 2014; Bekas *et al.*, 2016) and the adhesive can be detached without leaving a trace. Furthermore, new smart adhesive materials such as self-healing adhesive material, dis-bond adhesive materials and the ones with self-healing properties improve the durability of the structures and lower the costs (Blaiszik *et al.*, 2009). However, the research in the application of self-healing materials to adhesive joints is in the initial stages.

The dicyclopentadiene (DCPD) Grubbs catalyst self-healing system, in microcapsules, can be incorporated into the bulk of epoxy matrices used for the manufacturing of fiber-reinforced composites (Jin *et al.*, 2011; Aïssa *et al.*, 2012; Jones *et al.*, 2015) increasing the fracture toughness of the polymer (Blaiszik *et al.*, 2009; Zhang and Yang, 2014; Jin *et al.*, 2012). The epoxy can also be encapsulated like healing agent (Aïssa *et al.*, 2012; da Silva *et al.*, 2011) or dual microcapsule epoxy amine chemist as self-healing in thermoset epoxy (Saldanha *et al.*, 2013).

Crash resistant adhesives with high toughness and impact resistance as well as thermosetting resins as the conventional epoxies represent an interesting issue particularly relevant for the automotive industry (Imanaka *et al.*, 2009). Good results were obtained by adding polyurethane and rubber particles to epoxy adhesives (Imanaka *et al.*, 2010; Bartlett and Crosby, 2014; Dos Santos *et al.*, 2016). Of course, the environmental aspect is nowadays not negligible leading to consider natural adhesives (bio-adhesives) which can satisfy issues such as easy release and reusability (Zhang *et al.*, 2015). For example, water-based adhesive (Hermiati *et al.*, 2015; Wayakron Phetphaisit *et al.*, 2013), natural rubber (Li *et al.*, 2016; Brunner *et al.*, 2009) and modified soybean-flour (MSF) adhesives (Brunner *et al.*, 2013), are considered for wood composite materials and not for metal and CFRP based joints.

The adhesive material properties can be estimated using different ASTM and ISO standard test methods (Reis *et al.*, 2015; Renart *et al.*, 2016; Chaves *et al.*, 2014; Oliver and Johnson, 2009; Korta *et al.*, 2015; Li *et al.*, 2012; Mubashar *et al.*, 2011; Markatos *et al.*, 2013) (Fig. 4). However, given some shortcomings still to be overpassed, automated procedures are also proposed, for example with regard to Mode I and Mode II fracture.

In addition to the moisture absorption, the main environmental threats for adhesive joints are related to the effect of temperature on their strength and durability. This latter aspect is affected by fire and UV (ultraviolet) radiation, too.

The adherends

Different materials behave differently and affect the final performance of the joints. For this reason, the adherends should be selected with care. A significant difference in strength, for example, was observed, in the case of joints bonded with different adherend materials (Korta *et al.*, 2015; Li *et al.*, 2012; Mubashar *et al.*, 2011; Markatos *et al.*, 2013).

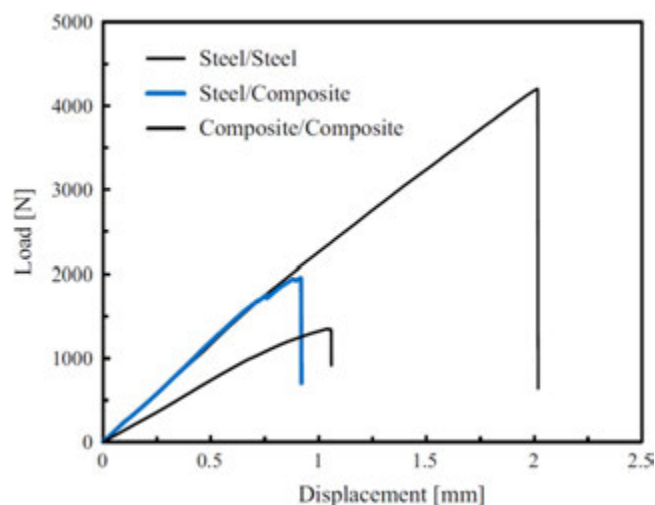


Fig. 4 Typical load–displacement curves for single lap joints with dissimilar adherends. Reproduced from Reis, P.N.B., Soares, J.R.L., Pereira, A.M., Ferreira, J.A.M., 2015. Theoretical and Applied Fracture Mechanics 80, 79–86.

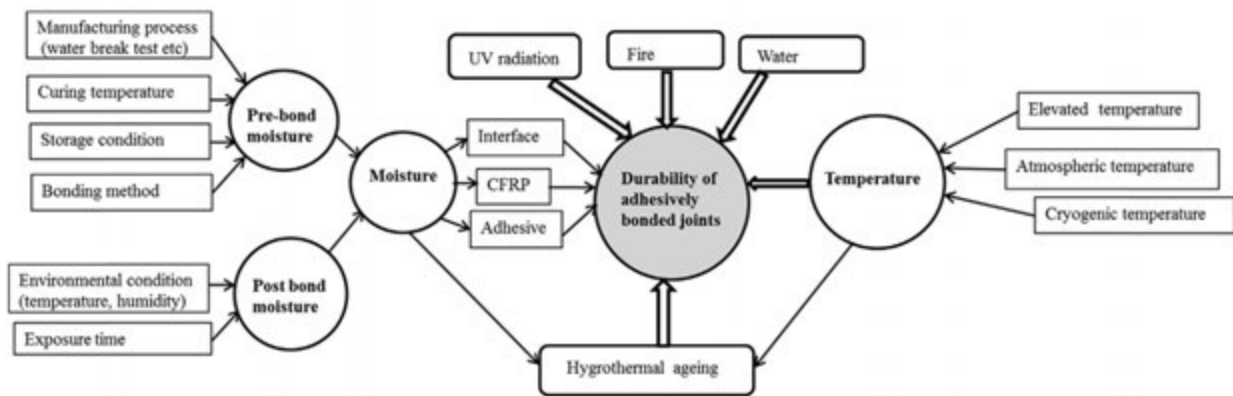


Fig. 5 Diagram of environmental parameters influencing the durability of adhesively bonded joints. Reproduced from Budhe, S., Banea, M.D., de Barros, S., da Silva, L.F.M., 2017. *International Journal of Adhesion and Adhesives* 72 (October), 30–42.

The compatibility of fibers and resin and their crucial role in the long-term performance of composite materials represent further aspects of fundamental importance. In general, there should be a good compatibility between the adherend and adhesive material in both chemical and mechanical terms.

Pre-bond and post bond moisture

In general, moisture can change the adhesive through plasticization, swelling, increase of cracks, hydrolysis, and reducing its glass transition temperature (Fig. 5). On the other hand, moisture is known to increase the ductility and to reduce the elastic modulus and strength of resin/adhesive even if, in cyclic moisture exposure condition, the ductility and the loss modulus are however compromised (Mohan *et al.*, 2013).

The effect of moisture content in the substrate before bonding on the mechanical performance of the joints is worthy of attention. The substrates absorb moisture before bonding mainly during the manufacturing process, storing the laminate for an extended period and during manufacturing bonded joints and this behavior affects a lot the final mechanical properties, leading to joint strength reductions (Budhe *et al.*, 2014; Pantelakis and Tserpes, 2014; Markatos *et al.*, 2014; Tserpes *et al.*, 2014; Stazi *et al.*, 2015; Mohan *et al.*, 2014) even if its effect on the composite structure is not well documented.

In general, a small amount of pre-bond moisture seems to have a positive or almost zero influence on the fracture toughness (Markatos *et al.*, 2014; Tserpes *et al.*, 2014; Stazi *et al.*, 2015) of bonded joints probably due to matrix ductility and toughness of the adhesive. However, wet substrates influence the failure mode, change the cohesive failure to interface failure and also promote multiple crack failure (Budhe *et al.*, 2014; Markatos *et al.*, 2014; Stazi *et al.*, 2015). Drying temperature and time led to the maximum performance of the joint. This area needs further attention even considering that the effect of pre-bond moisture on the fatigue behavior is still not well developed. It is well known that the adhesive and the resin matrix are the most affected by moisture in composite bonded structures. The moisture affects adhesive material, adherend material and bonding method by (1) altering the resin matrix; (2) damaging the fiber/matrix interface; as well as (3) influencing processing parameters as fiber-level degradation exposure condition and time, curing temperature, and so on (Chaves *et al.*, 2014; Carbas *et al.*, 2018; Zhang *et al.*, 2014; Sugita *et al.*, 2010; Costa *et al.*, 2018; Budhe, 2014; Sciolti *et al.*, 2010). The literature reveals that the degradation of the interface and weakening of intermolecular adhesive forces are often significantly more incisive than that of the adhesive (Markatos *et al.*, 2013; Mohan *et al.*, 2013; Pantelakis and Tserpes, 2014). Moreover, it was demonstrated that drying a substrate already bonded with wet adhesive materials can promote an increase in fracture toughness of the joint but does not provide complete recovery of its performances (Zafar *et al.*, 2012).

The moisture, by swelling the matrices, causes stresses large enough to pull the matrix away from the fiber and damages the fiber matrix interface. Hence, the properties dominated by the resin, such as interlaminar shear strength, are more susceptible to moisture-induced degradation than the fiber-dominated properties, such as tensile strength (Carra and Carvelli, 2014).

However, at present, it is not possible to define a clear correlation between moisture content and property degradation due to the simultaneous occurrence of several harmful mechanisms at the fiber/matrix interface. Besides, the fiber orientation and type and the resin matrix are other influential factors.

Recently, much interest has been dedicated to the environmental conditions at which adhesive joints are typically exposed, and most of the researchers (Korta *et al.*, 2015; Mohan *et al.*, 2013; Pantelakis and Tserpes, 2014; Mohan *et al.*, 2014) verified that post-bonding moisture adversely affect the joint strength and the extent of degradations depend on the exposure time, type of adherend-adhesive materials, manufacturing processes adopted, ageing conditions and specimen configuration. These drawbacks were mainly explained in terms of adhesive plasticization and weak interfacial failure (Mohan *et al.*, 2015). Drying resulted in being the best-suited treatment to recover the strength. Thus, the use of a proper drying process along with the careful selection of adhesive and polymer composite are the most appropriate strategies to alleviate these drawbacks (Mohan *et al.*, 2015; Banea *et al.*, 2014a; Heshmati *et al.*, 2015).

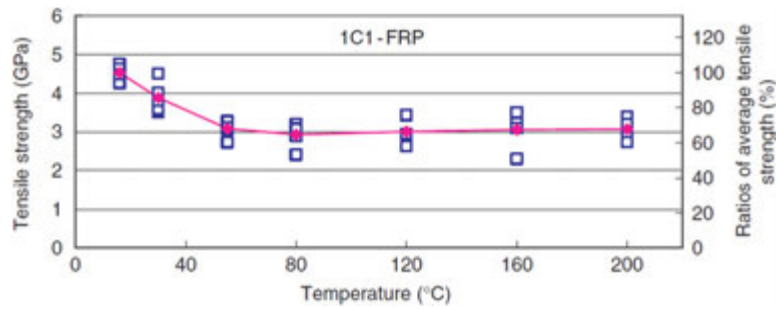


Fig. 6 Tensile strengths and average strength ratios of CFRP sheets at different temperatures. Reproduced from Cao, S., Wu, Z., Wang, X., 2009. *Journal of Composite Materials* 43 (4), 315–330.

The temperature

Given the polymeric nature of adhesives, the most critical factor in the design of a bonded joint is the variation of adhesive mechanical properties with temperature: stress-strain behavior and toughness. The main factors that determine the strength of an adhesive joint when used over a wide temperature range are the cure shrinkage, the coefficients of thermal expansion (CTE), and variations of their mechanical properties with temperature (Banea *et al.*, 2014a; Banea and da Silva 2009a; Banea and Da Silva, 2010; Banea *et al.*, 2012). In general, studies show a decrease in strength with both increasing and decreasing temperatures, see Fig. 6 (Wang *et al.*, 2011; Cao *et al.*, 2009; Nardone *et al.*, 2012; Hu *et al.*, 2013; Banea *et al.*, 2011b; Grant *et al.*, 2009). This drawback is due to the low adhesive strength at high temperatures and the high thermal stresses and the brittleness of the adhesive at low temperatures.

For temperatures higher than the glass transition, effects of the polymer matrix of the composite to be joined and/or of the adhesives on the viscoelastic response were demonstrated, but the fibers do not undergo any degradation (Hollaway, 2010). In particular, it was shown that exposures below glass transition temperature are advantageous for FRP composites and adhesives as a result of further post-curing (Wang *et al.*, 2011; Hollaway, 2010; Robert *et al.*, 2010; Wu and Yan, 2011).

Fluctuating temperatures can also lead to the progressive debonding and weakening of the materials and the fiber/matrix interface (Wu and Yan, 2011) due to the different thermal expansion coefficients of fibers and resin, highlighting the importance of their compatibility and its crucial role in the longterm performance of composite materials subjected to cyclic freeze/thaw loading.

Extreme low temperatures cause embrittlement, hardening and micro-cracking of polymer matrices and fiber/matrix bond degradations (Budhe *et al.*, 2014; Markatos *et al.*, 2014; Charalambous *et al.*, 2015).

Temperature affects the fracture behavior of the joints: the mode I toughness, G_{IC} , increases with increasing temperature with slight reductions detected below the room temperature (Li *et al.*, 2016; Coronado *et al.*, 2012; Im *et al.*, 2014; Banea *et al.*, 2011a). This behavior can be ascribed to increments of the matrix ductility, increase in fiber bridging and fiber breakage (Li *et al.*, 2016; Im *et al.*, 2014; Banea *et al.*, 2011a). The brittle behavior of the matrix at low temperature is responsible for low G_{IC} values (Banea *et al.*, 2011a).

On the other side, G_{IIC} decreases with increasing temperature (Coronado *et al.*, 2012; Park *et al.*, 2010; Zhang *et al.*, 2010) because of a reduced toughness of the fiber/matrix interface (Coronado *et al.*, 2012).

It is essential to observe that the combined effect of moisture and temperature through so-called hydrothermal conditions is more critical than the adverse effect of each specific condition (Carbas *et al.*, 2018; Budhe, 2014; Alessi *et al.*, 2014; Liu *et al.*, 2016a; Meng *et al.*, 2015; Viana *et al.*, 2017a,b).

The moisture absorption sensitivity increases at high temperature, reducing the resistance to the damage since the shear strength decreases (Liu *et al.*, 2016a).

Failure Modes of Adhesive Joints

The almost zero impact of the adhesive on the overall weight of the structure allows a particularly uniform load and, therefore, stress distribution, consequently improving its resistance to bending, fatigue and vibrations. Moreover, the adhesive joints provide smooth contours and do not alter the part dimension. However, one of the major limiting factors of this type of joints is the lack of quantitative non-destructive techniques that can detect defects in the adhesive bond.

Adhesive bond defects fall into three broad categories: gross defects, cohesive defects and adhesive defects. It is commonly accepted that *gross defects* such as cracking, debonding, voids and porosity can be detected using standard ultrasonic inspection techniques.

Poor cohesion is a degradation of the strength and other mechanical properties of the adhesive and can be caused at manufacture by curing errors or in service by an environmental attack. This strength degradation is commonly seen as a change in the elastic modulus of the adhesive and so cohesive defects can often be inferred from ultrasonic velocity measurements. Thus, cohesive failures occur when the adhesive joints are subjected to load exceeding the adhesive strength or the adherend strength. The former are localized effects occurring near stress concentrations while the latter failure mode generally initiates from the matrix between layers as a result of out-of-plane tensile or interlaminar shear stresses or, furthermore, of through-thickness tensile cracking.

Poor adhesion, instead, is a reduction in the strength of the adhesive/adherend interface. This type of failure, generally determined by either inadequate surface treatment or material mismatch, is not easy to be detected by conventional ultrasonic techniques because the bond strength is governed by a thin layer with a thickness many orders of magnitude less than an ultrasonic wavelength.

However, it is worth to note that, in the most severe cases of poor adhesion (*kissing bonds*), the adhesive joint can have such a low resistance that it is possible to obtain information even using ultrasonic methods.

Changes in failure modes depend on the moisture percentage content in the joints (Croxford *et al.*, 2018; Joseph *et al.*, 2018b). Moreover, also with the same materials and other parameters, the joint can fail in different ways depending on the joining method adopted such as co-bonded, co-cured and secondary bonding (Pantelakis and Tserpes, 2014; Heshmati *et al.*, 2015).

Mechanical Joints

Mechanical joints, realised by drilling holes in which fasteners can be installed, are widely used in composite structure assembly due to many peculiarities such as:

- (1) Cost efficiency and accessibility;
- (2) Possibility of obtaining repeated assembly and disassembly for repair and maintenance operations;
- (3) Easy inspection and quality control; and
- (4) Little or no surface preparation than adhesively bonded joints.

Among universal mechanical joints, bolted joints, constructed by using either a double or single-lap joint arrangement, remain the dominant fastening mechanisms in most applications, in particular in the aerospace one because of ease to assemble and disassemble (Joseph *et al.*, 2018a). Mechanical fasteners offer the advantage of being able to be removed without destroying the structure but have to be sized appropriately for structural optimization and its design represents a matter of concern (Joseph *et al.*, 2018a; Khashaba *et al.*, 2019; Camanho and Matthews, 1997; Thoppul *et al.*, 2009). The selection of appropriate and optimum geometric parameters and materials are necessary to achieve the structural integrity and reliability in composite structures.

Bolted joints in composites, in fact, can fail in loading conditions not predicted by either perfectly elastic or entirely plastic assumptions. Four basic modes of composite bolted joints including bearing failure, shear-out failure, net-section tension failure and bolt fracture were studied (Hazimeh *et al.*, 2015; Pramanik *et al.*, 2017; Aktas and Dirikolu, 2003). Since the net-section tension, the shear-out failure and the bolt fracture of bolted joints are crucial, bolted joints are usually designed to fail in bolt bearing.

The bearing strength of fiber-reinforced composite laminates is such a vital design factor. Currently, there is no unambiguous consensus on which method and which type of failure modes should be used to predict the strength of such joints (Hazimeh *et al.*, 2015).

Many studies in this area have used experimental data and numerical models to determine the optimum joint strength, predict the failure mode, and construct highly stable structures.

Unlike metals, fiber-reinforced polymers are linearly elastic up to failure, and consequently no yielding alleviates stress concentrations. Previous investigations demonstrated that slippage between resin and fibers averages out local stress concentrations and results in more of the fibers being loaded before final failure. This leads to the surprising conclusion that, in the vicinity of holes, improving adhesion between resin and fibers can, in fact, lower joint strength.

The effectiveness of this type of joints depends on the quality of the hole because the load that is transmitted via fasteners can lead to a stress concentration around the hole fastener boundary and may lead to the premature failure of structures. In other words, the drilling represents a very crucial point since composite materials, for their anisotropic nature, are very difficult to be machined without causing any delamination. This, together with the presence of fasteners that represents a discontinuity, lead to a stress concentration reducing the mechanical resistance of the joint. Moreover, other important aspects to be accounted for are the different thermal expansion of the fasteners and the necessary materials and the additional weight due to the presence of fasteners minimizing the weight-saving potential of composite structures.

In light of these premises, due to the complex nature of composite materials, many studies have been extensively conducted to understand the effect of factors as stacking sequence (Hart-Smith, 1980), fiber orientation (Lee *et al.*, 2015), joint geometry (size and distance between holes) (Achintha and Zirbo, 2018), clearance between the hole and pin, types of fasteners, temperature, moisture, clamping pressure and lateral constraints (Liu *et al.*, 2016b; Ascione *et al.*, 2009) on the strength of mechanical joints.

Hart-Smith (1980) showed that fiber composites with bolted connections might fail due to tension, shear-out, or bearing (Fig. 7). It may also fail by cleavage of the laminate, the connector pulling through the laminate, or by bolt failure either by single or double shear.

Another aspect recently studied is the hole perpendicularity error: a manufacturing defect that causes stress concentrations around the hole (Jeevi *et al.*, 2019).

Ascione *et al.* (2009) instead, studied the effects of the fiber inclination angle and laminate stacking sequence on the bearing failure load of composite bolted joints. Their experimental results reveal that the bearing load failure significantly depends on the fiber inclination angle to the external load direction and that the stacking sequence is not significantly related to the bearing failure.

Other typical fasteners involve:

- (1) *rivets*: usually suitable for joining laminates up to 3 mm thick. The closing pressure is not always readily controllable, resulting in a wide variation in clamping pressure. Also, the riveting operation can potentially damage the laminates; and

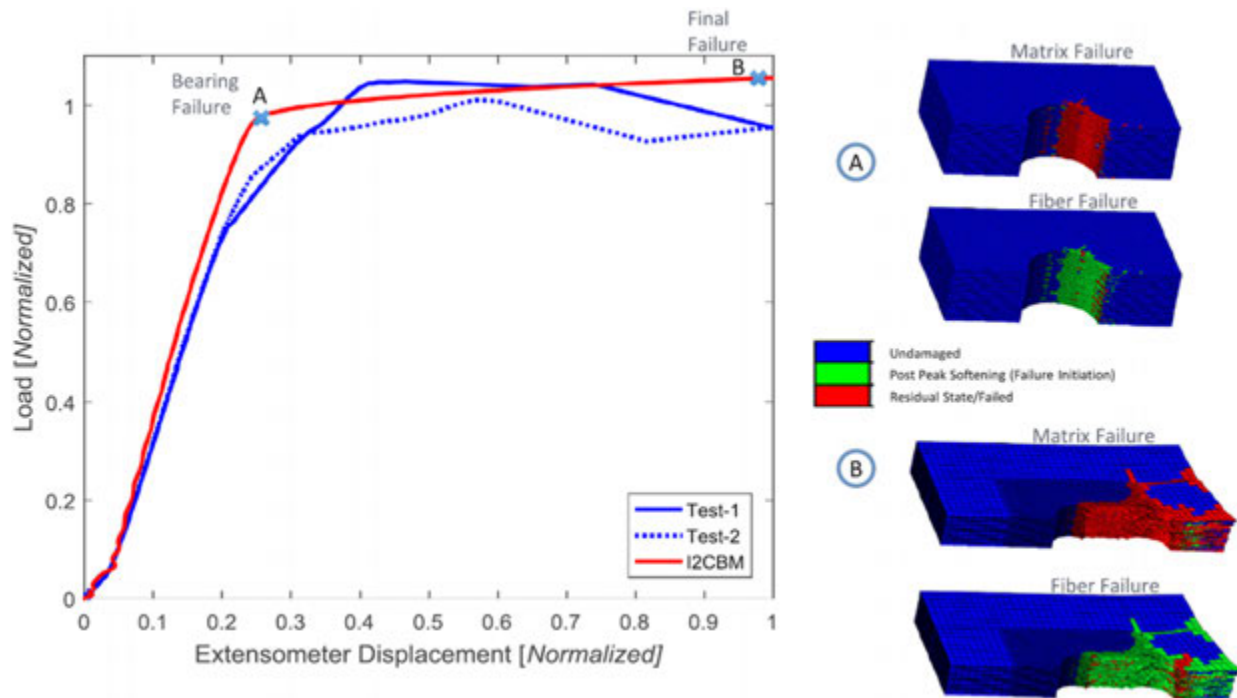


Fig. 7 Load-displacement response of double lap shear bolted joint and bearing failure status at critical points. Reproduced from Joseph, A.P.K., Davidson, P., Waas, A.M., 2018b. *Composites Part A: Applied Science and Manufacturing* 113, 264–274.

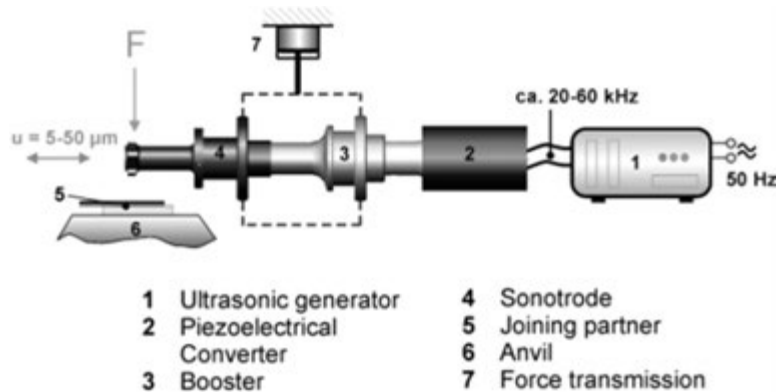


Fig. 8 Principle of the ultrasonic spot welding technique. Reproduced from Balle, F., Wagner, G., Eifler, D., 2007. *Materialwissenschaft und Werkstofftechnik* 38 (11), 934–938.

- (2) *pin joints*: this technique provides relevant potential to join different materials in terms of weight advantage since the overlapping length can be reduced and superior overall strength of the joint since the lower notch stress-induced concerning riveting and bolts joining. The pin junction allows moving the FRP fibers around the junction element avoiding their usual destruction in the preliminary drilling operation.

Welded Joints

Recently, a growing attention is dedicated to the development and application of thermoplastic composites given their intrinsic recyclability and improved damage resistance with respect to thermoset based counterparts.

In this case, the ability of thermoplastics to be re-melted after they are formed has risen an outstanding interest towards welding (fusion-bonding) techniques to joint unreinforced and fiber reinforced components based on these materials.

Welded joining methodologies can be roughly classified in frictional heating (e.g., ultrasonic welding), electromagnetic heating (e.g., induction welding), bulk heating (e.g., dual resin bonding) and thermal techniques (e.g., infrared welding, laser welding).

Below a brief outline of the basic principles and potentials of some welding processes developed over the last few decades is reported.

The *ultrasonic welding* (UW) is an industrial solid-state process consisting in the application of high-frequency (15–70 kHz) ultrasonic acoustic vibrations to induce local heating, under pressure, of plastic and/or metal components (**Fig. 8**). Expectedly, the efficiency of this method depends on many factors such as the physical properties of the materials to be joined, the frequency and amplitude of the ultrasonic waves and the geometric aspects of the joint (*Balle et al., 2007; Liu et al., 2001*).

The welding set-up involves an electrical generator of controlled high frequency current that is converted into mechanical energy through a transducer, an amplifier and a sonotrode that transmits the vibration to the piece to be assembled. The heat generated by vibrations and a pressure applied to the workpieces ensure the welding.

This joining process exhibits several advantages. First of all, avoiding the liquid-solid transformation it is a clean and environmentally friendly process. Other noteworthy strengths are: the easy automation and integration of the process in the production lines, the relatively low temperatures necessary for the welding of the components (in the case of pieces based on thermoplastic and thermosetting polymers/composites, it is sufficient to heat them to temperatures slightly above the material glass transition one), the possibility of electronically monitoring the welding parameters for subsequent evaluations in addition to very short welding times (*Ageorges et al., 2001*).

However, research efforts already available in the literature highlight, among other things, the need to have surface asperities able to function as energy directors to focus the heating at the interface and, therefore, to enhance the mechanical performances of the junction obtained.

Currently, only a few studies are available regarding the application of the UW process for the junction of thermoplastic FRP (*Liu et al., 2001; Li et al., 2004; Benatar and Gutowski, 1989*).

Among these, according to Benatar and Gutowski (*Villegas et al., 2013*) the UW process takes place into 5 stages that are closely interconnected: vibration of the parts, viscoelastic heating of the thermoplastic matrix, heat transfer, localized wetting of the interfacial region and intermolecular diffusion of the polymeric phases.

The *induction welding* (IW) (*Kagan and Nichols, 2005; Stokes, 2003; Ahmed et al., 2006; Villegas et al., 2013*) exploits the heat generated by induction using a radio-frequency alternating current that magnetically excite an implant placed at the interface of parts to be joined. Thus, the welding set-up consists of a generator of electrical energy that powers a high frequency induction coil, in turn, designed to generate a high-frequency electromagnetic field. If the interested surfaces to be assembled are surrounded by electro conductive and/or magnetic susceptible materials, eddy currents, with a frequency equal to that of the electromagnetic field, will heat the pieces by Joule effect.

This joining method, widely used for the production of aeronautical assemblies, allows to obtain high-quality, pressure-tight structural welds with most thermoplastic FRPs in just a few seconds. The addition of interfacial implants ensures the obtainment of highly efficient joints, even in presence of surface irregularities or large tolerances between the parts (**Fig. 9**).

Main parameters influencing the inductive heating are the coupling distance, the field frequency, the coil current and the inductor geometry (*Ochinero and Hyer, 2002*).

Although this methodology has been limited, for a long time, to flat joints, recent researches have developed a three-dimensional process able to join complex and curved parts with a high degree of automation.

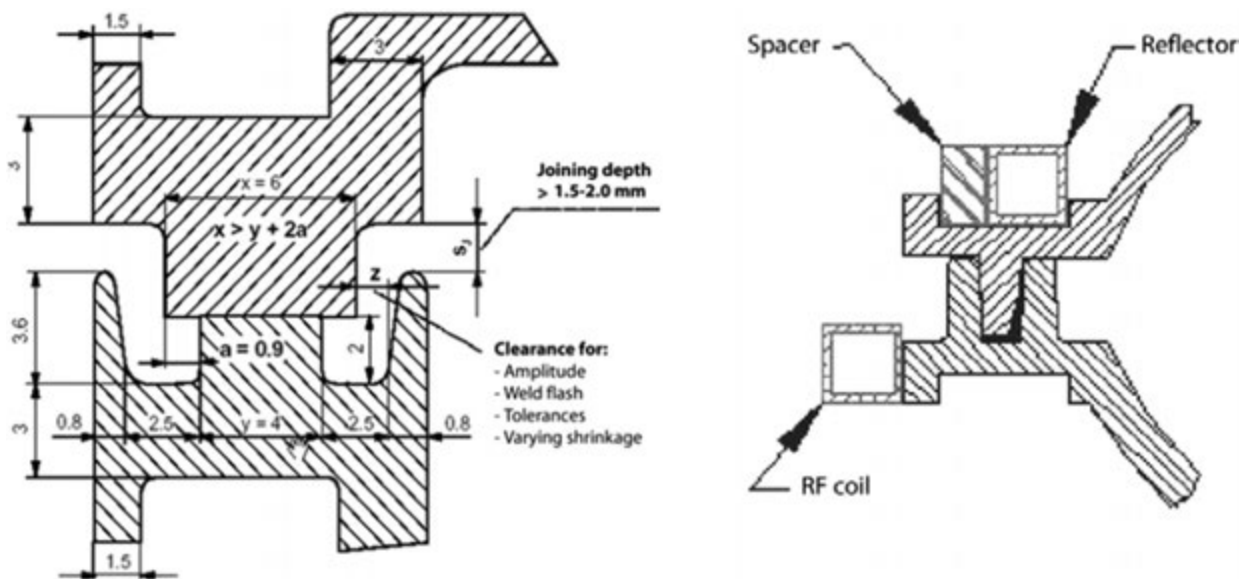


Fig. 9 Schematic views of (linear vibration welding) butt-joint and (induction welding) tongue-in-groove joint. Reproduced from Kagan, V.A., Nichols, R.J., 2005. *Journal of Reinforced Plastics and Composites* 24 (13), 1345–1352.

The *Microwave welding* (MW) (Fernandez Villegas and Vizcaino Rubio, 2015; Choudhury and Debnath, 2019; Hashmi *et al.*, 2015; Bajpai *et al.*, 2013) process consists of the junction of materials, mostly with a thermoplastic matrix, which undergo dielectric heating by the simultaneous action of a high-frequency alternating electromagnetic field and an applied pressure between the surfaces to be joined. Unlike conventional heating, since the microwaves can penetrate the material, heat is simultaneously generated within the volume of the interested pieces through molecular interaction with the external electromagnetic field. Considering that normally plastics do not heat up due to microwave exposure, the process is usually favored by the use of high dielectric susceptor material as ceramics, metal, carbon or conducting polymers, located at the joint interface.

Main parameters affecting this joining method are exposure time, intensity and frequency of electromagnetic radiation, applied pressure, nature and quantity of susceptible material used at the interface.

Regarding the benefits of MW, instead, it is worth mentioning the speed of the process (minutes), the possibility of joining pieces of complex geometry, the uniformity of heat distribution within the materials, the limited environmental impact due to no generation of polluting gases and the homogeneous microstructure of obtained joints that generally exhibit a high strength.

Resistance welding (RW) (Dobrzański, 2019; Brassard *et al.*, 2019; Stavrov and Bersee, 2005) is a joining process by which the heat between the parts is generated by the passage of an electric current, in a conductive layer (e.g., stainless steel mesh, carbon fibers) positioned at the interface of the same, for a predetermined time. The interface heating, generated in accordance with the Joule-Lenz law, merges the plastic phase of the parts to be joined and, thanks also to the action of a pressure applied between the contact surfaces, the pieces consolidate to form a unique component.

These joining methodologies can be classified according to the shape of the workpieces and the form of electrodes in several variants among which it is worth to mention: spot welding (Karami Pabandi *et al.*, 2017), projection welding, seam welding and butt welding.

In general, the most important parameter that affects the effectiveness of this process and the quality of the joints obtained is the current intensity applied since the amount of heat generated (Q) varies with the square of this factor:

$$Q = I^2 R t \text{ (Joule – Lenz law)}$$

where I is the current intensity, R is the resistance of the conductive layer at the interface and t is the time of the current flow.

Increasing the current intensity favors a greater welding speed but causes the electrodes deterioration. Similarly, the current flow time must be sufficient to ensure adequate welding but not excessively prolonged to damage the electrodes.

The contact pressure between the parts to be joined (welding force) must allow the passage of current but influences the process as it acts on the interfacial resistance and on the contact area for the deformation of the materials. If the welding force is too low, as the contact resistance is too high, a large amount of heat is generated which can cause immediate expulsion of the pieces after starting the welding current. If the welding force is high, however, the contact area will be large with consequent low current density and low contact resistance which will reduce heat generation and the size of the welding nugget (Fig. 10).

Other determining factors can be considered dimensions, geometry and electrical resistivity of the parts to be assembled, the contact resistance as well as the electrical and mechanical characteristics of the welding machine.

The *Laser Welding* (LW) (Labeas *et al.*, 2010; Knapp *et al.*, 2010; Jung *et al.*, 2013) method uses a highly concentrated beam of light to join metal or thermoplastic composite pieces. The excitation of the electrons in the area exposed to the intense beam of light (laser) induces the fusion of the materials and favors their junction.

In this process, unlike the previous one, no electrode is used and no form of tool wear occurs. Furthermore, laser welding is highly specific in targeting and generates high quality welds.

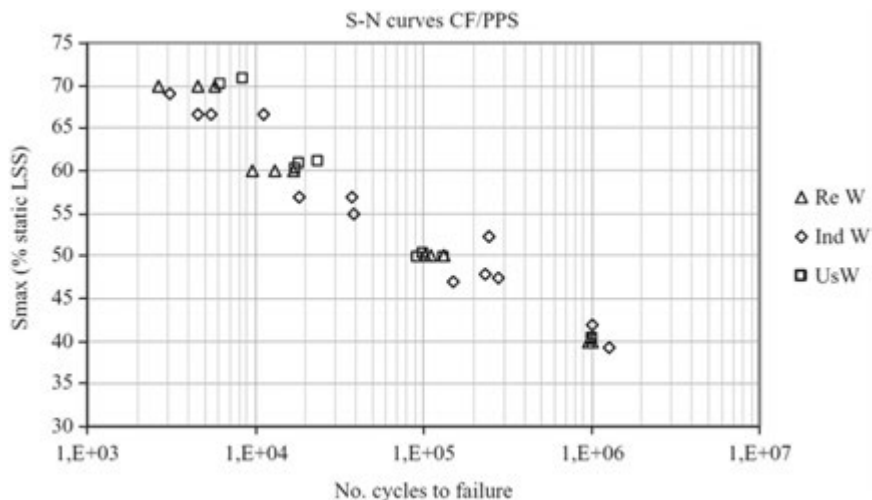


Fig. 10 S-N fatigue data for resistance, induction and ultrasonically welded samples. Reproduced from Villegas, I.F., Moser, L., Yousefpour, A., Mitschang, P., Bersee, H.E.N., 2013. *Journal of Thermoplastic Composite Materials* 26 (8), 1007–1024.

Among noteworthy parameters influencing the effectiveness of the process and the quality (microstructure/strength) of joints are laser power, welding speed, focal position as well as nature of materials to be joined.

Hybrid Joints

Joints obtained by combining two or more joining operations carried out simultaneously or sequentially intending to exploit any synergistic load bearing interaction in service conditions (ASTM International, 2014). Typical hybrid joints include an adhesive in combination with a mechanical fastener or spot weld. An advantage of this strategy is, for example, that the mechanical junction, with immediate effect, can fix the position of the components until the polymerization of the adhesive takes place, facilitating the assembly process.

In general, it is possible to combine different mechanical joining, different adhesive bonding, mechanical/fusion welding methods, and so on. For example, an important group of hybrid joining techniques, commonly used in structural applications, involves the combination of two different fusion welding methods and allows of exploiting the advantages of individual processes.

Mechanical Testing of Joints

Several standards exist to determine the strength of a joined composite material since composite laminates can be with continuous or discontinuous-fiber (tape or fabric), constituted by balanced and symmetric laminates concerning the test direction, or involving different architectures, such as woven fabric or unsymmetrical laminates.

The open hole tensile (OHT) strength or the notched tensile strength of polymer-matrix composites is useful to understand the strength necessary to know for structural design allowable, material specifications, and research and development. The ultimate strength is affected by factors such as bolt-hole preparation, specimen geometry, and thickness scaling. For this reason, ASTM standard test method D 5766/5766M-02a (ASTM International, 2014) suggests the test specimen geometry.

Open hole compressive (OHC) tests can be performed following the suggestions from the standard test method ASTM D 6484/6484M-04. The load is transferred through shear to the specimens. The specimen geometry is similar to that used for OHT (ASTM International, 2014). Ultimate OHC strength is determined only if the failure modes passes through the bolt hole.

The bearing strength in tension and compression loading, defined as the bearing stress developed when the bolt-hole is deformed by 4% of the initial bolt-hole diameter, is determined according to the ASTM D 953-02 (Bearing strength of rigid plastics).

The ASTM D 5961/D 5961M-05 standard test method is, instead, adopted to determine the bearing response of polymer-matrix composite laminates in double-shear tensile loading or single-shear tensile or compressive loading (recommendations by the military handbook MIL-HDBK-17F working group US Department of Defense, 2002; Shyprykevich, 1995). In a double-shear test, the bearing load is applied by pulling the specimen in tension through a torque of about 2.2–3.4 Nm of a fastener or pin with $D = 6$ mm. The effective bearing strain and the effective bearing stress are calculated by monitoring the applied load and the bolt-hole deformation during the maximum load is reached. In this case, both the load carried by the specimen at failure and the maximum load by the test specimen before the failure are recorded.

The loads are also recorded in the single-shear test, and the ultimate bearing strength is calculated. The test is carried out by pulling the torqued specimens. Two identical specimens similar to the specimen used for double-shear are fastened together through one hole or two holes located centrally near one end by a single shear, single-fastener test or a single-shear, double-fastener test, respectively. The bearing response is affected by the difference between hole and fastener diameters.

Bearing chord stiffness, E_{br} , ultimate bearing strength, F_{br} , and the offset bearing strength, F_{br} , are evaluated from the bearing stress/bearing strain plot. In particular, the first parameter is calculated between two specific bearing stress or bearing strain points on the linear portion of the curve from the changes in the bearing stress and the bearing strain over the chord stiffness range. The ultimate strength and the offset bearing strength (using 2% bearing strain) are calculated after correcting the bearing stress/strain data for the new effective origin. ASTM D 6873-03 and D 5961/D 5961M standards can be used to determine the bearing fatigue behavior of composite materials subjected to cyclic bearing loads in both double shears with single-fastener single-shear in single- or double-fastener configurations.

Bearing fatigue response of mechanically fastened composite joints is affected by fastener selection, fastener preload/torque, fastener bolt-hole clearance, environmental conditions, specimen geometry, support fixtures and test configuration. At this regard, measurements of the temperature of the specimen and the fastener during the test are suggested. High loading frequencies, indeed, may cause significant temperature rises and corresponding variations of the composite specimen properties.

The shear load transferred by a particular fastener is the bearing load, while any remaining load gets transferred or is bypassed through other fasteners. Various test methods and fixtures are available (Crews and Naik, 1986) to obtain the bearing/by-pass interaction response categorized as (1) independent bolt load (2) passive and (3) coupled bolt load/by-pass load. The ASTM D 7248/D 7248M standard for bearing/by-pass interaction response of polymer laminates is used to determine the bearing portion of by-pass interaction in composite bolted joints, where the bearing load is measured independently. The procedures for these methods are given by the MIL-HDBK-17F (Crews and Naik, 1986) and the recommendation by NASA Langley Research Center. Due to the weak strength of composite structures in the transverse direction, the bearing/bypass

interaction response, when the joint is subjected to transverse bearing loads independent of longitudinal by-pass loads, has to be known. In particular, it is important to know the pull-through strength that is the maximum load a mechanically fastened composite plate can sustain when the composite plates are pulled-apart perpendicular to the plane of the plates. ASTM D7332/D7332M-07 or the fastener pull through strength test method give suggestions to determine the pull-through strength of a composite plate/fastener combination and to evaluate different components of the fastener (bolt/nuts, pin/collars or washers). At this regard, two methods, consistent with the procedure given in the military handbook working group MIL-HDBK-17F (Crews and Naik, 1986), are recommended on flat plate specimens having rectangular cross-sections with a circular hole at the center for the fastener. Useful results are obtained by Banbury and Kelly (1999) on carbon fiber epoxy prepreg with plain weave and unidirectional tape having a stacking sequence of $[0,90/\pm 45]_s$. Fasteners with protruding and countersunk heads in different diameters were finger tightened to the composite plate. Since the failure load increases at the increasing fastener radius and thickness (Banbury and Kelly, 1999), authors deduced that composite joints loaded in pull-through may fail in either matrix shear fracture from the outer edge of the fastener head or in flexural failure in lower and upper plies near the bolt-hole boundary.

Textile Composites

The standard test methods discussed above could not be appropriate for testing “textile” composite materials having continuous networks of braided, woven, knit, or stitched fibers, where the microstructures differ significantly from those of tape laminates. For example, under uniaxial tension inhomogeneous local displacements develop within the textile composites, not observed in case of laminates made of unidirectional prepreg tapes.

In the area of textile composites, standard methods for tensile tests (Peng *et al.*, 2012), and the development of standard test method to determine the bearing strength of textile composites (Kroeger, 2014) have been sponsored by NASA. Bearing strengths of textile composite joints made of 2D triaxial braids were measured under three tensile loading configurations: unstabilized single-shear, stabilised single shear and double-shear. Bearing strengths of 3D woven composite joints were using stabilised single-shear tensile tests only.

The bearing strengths in unstabilized single-shear tests are typically lower than those in the stabilised single-shear tests due to bending of the unstabilized shear specimen during loading – the double-shear test in which the bending is almost eliminated results in the highest bearing strength. Comparing the average bearing strengths of joints with different braided architecture, it is clear that the double-shear test configuration yields the highest strength and the unstabilized single-shear test configuration gives the lowest strength. The important observation is that the difference between different braided architecture is slight and small tow sizes and small braided angles may result in strength improvement.

Non-Destructive Evaluations of Joints

The actual development in engineering consists of the use of a mix of different materials. The multi-material needs joints to ensure a durable connection. Different materials are mostly joined adhesively where the surface interested need to be adequately prepared. Sometimes it is necessary to join the parts without the possibility to use adhesives bonding but by using elements like rivets and bolts paying attention to ensure the integrity of the materials and the connections. For the safety of the structures it is necessary to test the joints without harming the parts. Common structures show an increasing amount of mechanical joints often combined with adhesives. The performance of the joints can be compromised by the presence of defects that can appear during the manufacturing processes as well as during the service. From this the necessity of non-destructive techniques able to monitor the integrity of the structures in correspondence of the joints.

Numerous techniques are used for the non-destructive evaluation of composites (Table 1), including ultrasonic testing (Vavilov *et al.*, 2015) thermographic testing (Tan *et al.*, 2011), infrared thermography testing (Bossi and Giurgiutiu, 2015), radiographic testing (Sarasini and Santulli, 2013), visual testing (VT) or visual inspection (VI) (Su *et al.*, 2014), acoustic emission testing (AE) (Hung *et al.*, 2013), acoustic-ultrasonic (Liu *et al.*, 2014), shearography testing (Yang *et al.*, 2013), optical testing (Kalinichenko

Table 1 Suitability of non-destructive testing methods for carbon fiber components

	Fibre orientation	Laminate structure	Complex geometries	Measurement time	Handling	Additional material
Acoustic/ultrasonic	●	●	●	●	●	○
Optic	●	○	●	●	●	●
Thermal	●	●	●	●	●	●
Radiographic	●	●	●	○	●	●
Electromagnetic	●	●	●	●	●	●

Note: Berger, D., Zaiß, M., Lanza, G., *et al.*, 2018. Production Engineering 12 (2), 161–172.

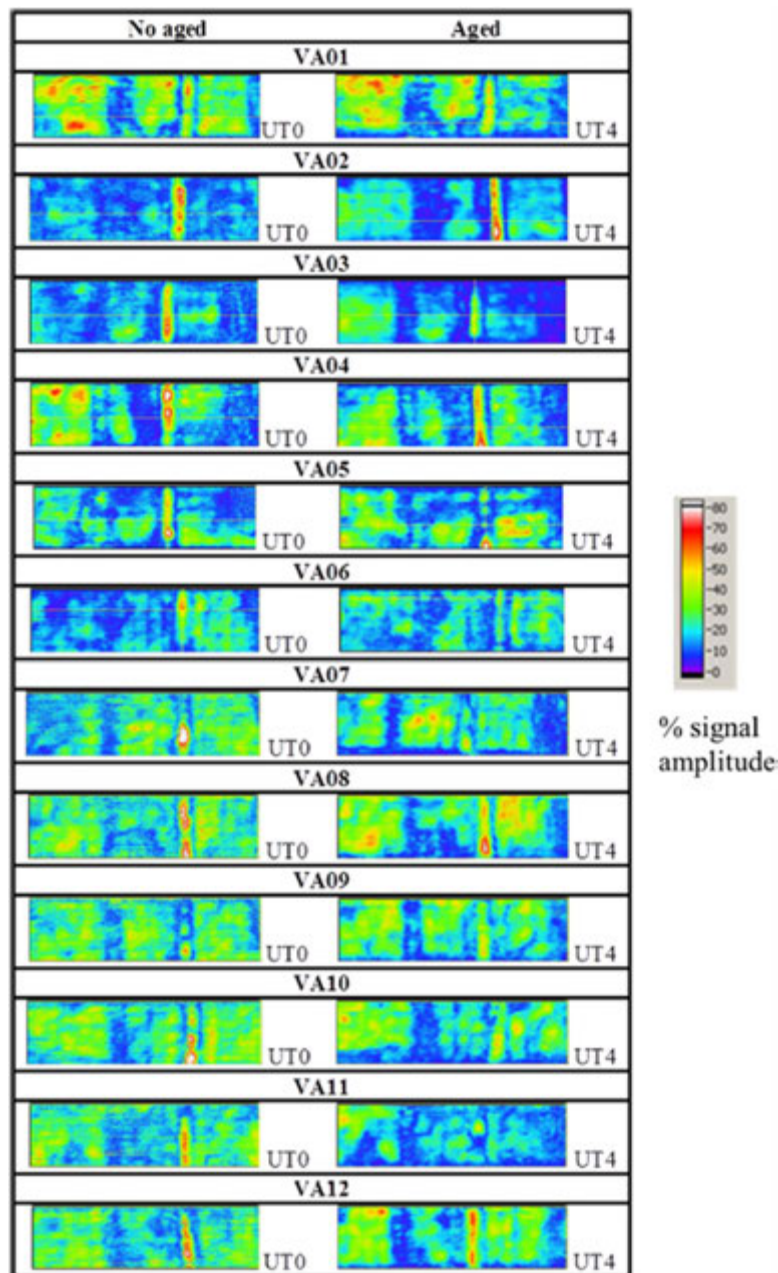


Fig. 11 UT images from C-scan of no aged (UT0) and aged (UT4) single-lap joint samples. Reproduced from Palumbo, D., Tamborrino, R., Galletti, U., *et al.*, 2016. NDT and E International 78, 1–9.

et al., 2013), electromagnetic testing (Lu *et al.*, 2013), liquid penetrant testing (Adams and Cawley, 1988), and magnetic particle testing (Mulaveesala *et al.*, 2013).

The basic types of NDT may be roughly distinguished in contact and non-contact methods. In particular, the former require good contact between the sensor and tested composite surface to obtain reliable data, but this may be a problem in the presence of mechanical joints. Contact methods are traditional ultrasonic testing, eddy current testing, magnetic testing, electromagnetic testing, and liquid penetrant testing. Non-contact methods, instead, are performed through transmission ultrasonic, radiography testing, thermography, shearography, optical methods (e.g., thermography, holography or shearography) and visual inspection (Hung *et al.*, 2013).

The choice of an ND analysis method rather than another will depend from time to time on the particular case and the particular type of joint, taking care to minimize costs, too.

Here follows a brief description of the methods most used for the analysis of joints based on polymer composites.

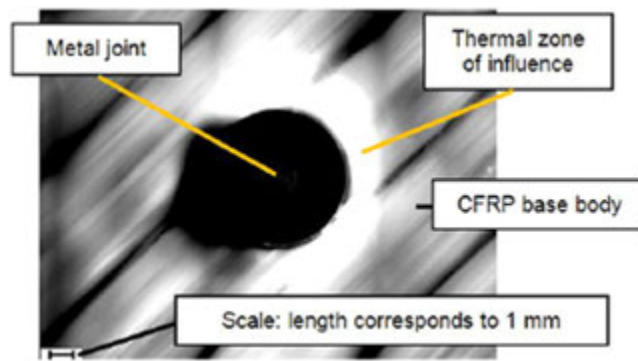


Fig. 12 Thermal zone of influence of a metal rivet in a CFRP base body leads to a lack of thermographic contrast (white area). Reproduced from Jelinek, M., Seidel, C., Reinhart, G., 2015. *Procedia CIRP* 37, 211–217.

Visual Testing (VT) – It is the primary type of NDT widely used due to its speed, economy and the relative reliability of the process. However, this method has an intrinsic disadvantage such as the expected high subjectivity.

Ultrasonic Testing (UT) – Method that utilizes sound waves mainly for detection of cracks and defects in parts and materials, dimensional measurements and more (Fig. 11).

A typical ultrasonic inspection apparatus involves:

- (1) An electronic device (pulse) producing high voltage electrical pulses;
- (2) A transducer is generating high frequency ultrasonic energy which is introduced and propagates in the form of waves into the examined materials; and
- (3) Display devices.

In the presence of defects, part of the ultrasonic energy is reflected from the flat surface. The reflected signal is transformed into an electrical one by the transducer, and its intensity is usually displayed versus the time from signal generation to when an echo was received. Given the velocity of the waves, the signal travel times can be directly related to the position of the defects.

Advantages of ultrasonic testing include speed of scan, good resolution and flaw detecting capabilities, superior depth of penetration for flaw detection or measurement with respect to other NDT methods. Disadvantages include the difficulty of set up, the examined surfaces must be accessible to transmit ultrasound, the need of a coupling medium to promote the transfer of sound energy into the test specimen and adequate skills to scan a part accurately.

The *Infrared Thermography Testing (IRT)*, based the thermal radiation emitted by a surface of a specimen and recorded by the thermal camera (Tan *et al.*, 2011), is considered a powerful technique in the inspection of composite materials due to the high emissivity of these materials, the non contact aspect, the fast inspection and easy interpretation (Fig. 12).

It is a non contact technique, but the problem in adhesively joined composite is that the presence of the adhesive, reducing heat fluctuations, could change the thermal conductivity of the substrate material. There are many advantages and disadvantages to this type of inspection. One of the advantages is that it can examine a large area of the part. Then, it does not have to be coupled on the surface, and it can be used for the inspection of parts with limited accessibility (e.g., only on one side). On the other hand, the high costs of infrared cameras and the need for specialized operators for the correct functioning of both the dedicated instrumentation and the acquisition system are aspects that can limit the use of this investigation technique.

Radiographic Testing (RT): there are many types of radiography, and each has specific applications. Conventional radiography is the most useful when the parts are neither too thick nor too thin. As a function of the thickness the voltage can be changed.

Gamma rays radiography is useful for thick parts because the gamma rays have shorter wavelengths.

There are varieties of radiographic testing methods for different applications. These methods are film radiography (Katunin *et al.*, 2015), computed radiography (Aidi *et al.*, 2015), computed tomography (Koyama *et al.*, 2013), and digital radiography (Hung *et al.*, 2013). Noteworthy advantages of tomography in comparison with the projection radiology are the 3-D visualized image of the structure and the rapid data processing. There are varieties of radiographic testing methods for different applications. These methods are film radiography (Katunin *et al.*, 2015) computed radiography (Aidi *et al.*, 2015), computed tomography (Koyama *et al.*, 2013), and digital radiography (Hung *et al.*, 2013). The great advantage of Tomography in comparison with the projection radiology is the 3-D visualized image of the structure, and the data is readable quickly and.

Electromagnetic Testing (ET) induces electric currents, magnetic fields, or both inside the material to test and observes the electromagnetic response. It includes Eddy Current Testing (EC) (Schroeder *et al.*, 2002), Remote Field Testing (RFT), Magnetic Flux Leakage (MFL) and Alternating Current Field Measurement (ACFM). In each of these techniques, the underlying physics is fundamentally different as the fields described by different classes of partial differential equations (PDEs).

Acoustic Emission (AE) is an effective method of imperfection analysis and could be used to evaluate the quality of a joint, in particular, for the adhesive ones. A combination method of acoustic and ultrasonic testing can be explicitly used to determine the severity of internal imperfections and inhomogeneity in a composite.

In nondestructive testing, the acoustic/ultrasonic class of testing has great potential based on optimal economy, flexibility and sensitivity. It allows to detect and analyse non-critical flaws, and it is a good indicator of accumulated irregularity in a structure. The disadvantage of this type of inspection is the setup.

Shearography Testing is a laser optical method. The discontinuity in a composite produces a stress concentration, and the criticality of defects will easily deduct by the degree of strain concentrations around a particular defect (Hung, 1989).

In the case of polymer composites, the nature of the joints is generally similar to that of the fundamental polymeric matrix, and this could limit the detection capacity of some techniques like for example IRT. The latter could present some difficulties from the weak contrast between the thermal properties of the constituents, the small dimensions of the thicknesses of the joints, and from the depth of the bonded interface. In the paper (Dawood and Rizkalla, 2010), a strategy was presented to design a new joint material adapted to the detection by using a numerical model: the epoxy resin was reinforced by a conductive boron nitride particles.

In (Schroeder et al., 2002), the authors proposed an advanced thermal NDT able to evaluate the quality of the joints in a composite pickup box with bonding: pulsed thermography. Excitation through a flash lamp capable of inspecting a large area seems to allow better repeatability, a more exceptional ability to detect defects in FRP composite joints, an increase in inspection speed, automation and non-contact capacity. The technique could be used to assess the initial state of the as-manufactured joint, monitor the performance of the parts over time and under various load conditions. Furthermore, the pulsed thermography, by monitoring the useful performance of the piece, can provide useful information for optimal design of the same. It has a concise cycle time (flash duration 5 ms) and can be used in real time, for example in the online production process for part validation.

Conclusive Remarks

The choice of the joint manufacturing method generally depends on the substrate to be bonded and field of application. However, there is a deficiency of understanding of bonding methods on the failure behavior and the evaluation of the mutual dependence between bulk and joint strength.

The existence of defects or discontinuities in the joints due to the different parameters is entirely undesirable for many applications. Mechanical investigations and non-destructive evaluation (NDT) play a fundamental role to evaluate the integrity of joints especially in case of dissimilar material assemblies.

The selection of the most appropriate method of investigation, obviously conditioned by the physical and geometrical characteristics of the parts to be assembled as well as by the actual conditions of use of the reference joints, often cannot disregard economic considerations.

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Joining of Polymer Matrix Composites Through Friction Stir Processes

VP Mahesh¹, Sooraj Patel¹, Anurag Gumaste¹, and Amit Arora, Advanced Materials Processing Research Group, Indian Institute of Technology Gandhinagar, Gandhinagar, Gujarat, India

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Glossary

Advancing side (AS) The side of the weld specimen where the tool rotation and the tool traverse is in the same direction.

Friction stir welding (FSW) A non-consumable rotating tool with a specially designed pin and shoulder is inserted into the abutting edges of sheets or plates to be joined and traversed along the line of joint.

Retreating side (RS) The side of the weld specimen where the tool rotation and the tool traverse is in the opposite direction.

Tool pin The probe section of the welding tool which is inserted into the workpiece to assist material flow and frictional heat generation.

Tool shoulder The section of the welding tool which takes part in the frictional heat generation when in contact with the workpiece.

Introduction

Polymer Matrix Composite (PMC) materials offer certain advantages over metallic materials such as high strength-to-weight ratio, low thermal conductivity, dampening, corrosion resistance, etc. Joining of similar and dissimilar PMCs is necessary to cater to strategic applications in the automobile, aerospace, and transportation industry. Adhesive bonding and mechanical fastening methods can be replaced by welding concepts such as friction stir welding (FSW), laser welding, ultrasonic welding, and resistance welding to produce sound joints. A comparison of the joint efficiency for polymer (PP) using different welding techniques is provided in [Fig. 1](#). FSW has become a potential candidate for the joining of thermoplastic polymers and polymer matrix composites ([Huang et al., 2017](#)).

Friction Stir Welding (FSW), a solid-state joining technique, offers several benefits over the conventional joining techniques. It was invented by The Welding Institute, UK in 1991. Primarily, FSW was focused to join high strength aerospace alloys. It has high industrial importance owing to the joining demands of the lightweight design structures. A specially designed tool having a rotating pin is inserted at the interface of the workpieces. The tool rotation produces viscoplastic deformation along with the frictional heating. Viscoplastic deformation of the base material takes place under an applied downward force by the shoulder. The welding of the workpiece is possible by the traverse motion of the tool in the welding direction. The localized heating due to friction softens the material, which further moves along with the rotating tool and forms a weld in solid-state condition. The schematic representation of the FSW process is shown in [Fig. 2](#).

Although FSW is primarily used for the joining of metallic materials, it has also been explored as a method to join polymers and polymer matrix composites. FSW has been used to join dissimilar materials such as metal to metal, metal to metal matrix composite (MMC), metal to polymer, polymer to polymer, and polymer to PMC materials ([Eslami et al., 2017](#)). The wide acceptance of FSW in industrial applications is attributed to the possibility of fully automated operation during the joining of materials. An added advantage of using FSW for Polymer Matrix Composites (PMC) is that the rotating tool additionally performs the task of mixing the reinforcement. This leads to the formation of a mutually intermeshed structure to uniformly distribute the reinforcement in the weld region ([Huang et al., 2017](#)).

The claim for FSW of polymers being a fully solid-state joining process can be contested. The smaller length molecules in polymers exhibit lower melting points and may undergo partial melting during FSW. As a result, inhomogeneous distribution is identified in the stirred zone which leads to deterioration of mechanical properties. Increasing the tool rotational speed to mix the material is not a preferred solution as the heat generation increases with an increase in rotational speed which further increases the partial melting of the base material ([Rezaee Hajideh et al., 2018](#)). The frictional heat generated during FSW of the polymer is considerably low as compared to the heat generated during the FSW of metals ([Eslami et al., 2015](#)).

In R&D laboratories a vertical milling machine can be used to join polymers and PMCs, while in industrial applications the operations can be handled by industrial robots. FSW can produce strong welds with better mechanical properties compared to other existing conventional joining techniques owing to the lower joining temperature. However, defect-free welds by FSW are still hard to obtain and various innovative tool designs (as discussed in section "Tool Design") have been proposed for welding of PMCs ([Eslami et al., 2017](#)). The FSW tool dimensions and design-features are key factors to the defect-free joining of PMCs. The joining of PMCs using friction stir based methods can be performed in various modes and configurations. Some of these are discussed in the section "Processes and Experimental Procedure". The tool design and welding process parameters impact the weld

¹Equally contributed author.

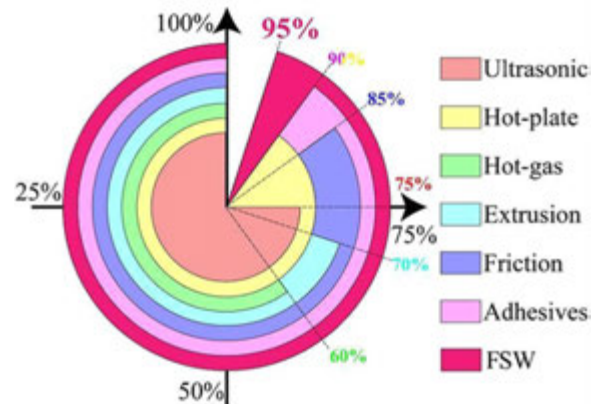


Fig. 1 Comparison of joint efficiency of PP welded using different techniques. Reproduced from Huang, Y., *et al.*, 2017. Friction stir welding/processing of polymers and polymer matrix composites. *Composites Part A: Applied Science and Manufacturing* 105 (2), 235–257.

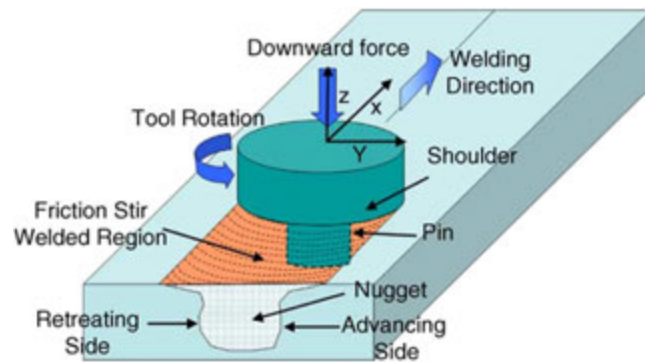


Fig. 2 Schematic of the friction stir welding process. Reproduced from Mishra, R.S., Ma, Z.Y., 2005. Friction stir welding and processing. *Materials Science and Engineering: R: Reports* 50 (1–2), 1–78.

properties. The importance of the tool geometry and process parameters during FSW of PMCs is discussed next in the section “Microstructure Evolution and Material Flow Analysis”.

Heat generation and material flow are the two most important physical processes during FSW. Understanding of these physical processes can help in the improvement of the process. The material flow analysis during FSW of PMC using microstructural evolution is discussed in section “Processing Parameters”. The mechanical properties such as tensile strength, shear bond strength, hardness, and impact strength for FSW of PMC are discussed in section “Mechanical Properties”. The common welding defects are presented in section “Defects” and several numerical studies of FSW of PMCs are shown in the section “Summary”.

Tool Design

The tools used for the FSW possess a tool pin and a tool shoulder. The tool pin and the tool shoulder are the sources of frictional heat generated during the welding. The design of the tool shoulder and the tool pin can greatly influence the weld quality. The tool profiles are varied based on the type of materials to be joined. Tool pin profiles determine the material flow during welding. The tool shoulder provides sufficient heat generation for the welding. The trailing edge of the tool facilitates the forging action during the tool traverse. **Fig. 3** shows the schematic of common tool pin profiles used for the friction stir welding. Different tool pin profiles such as cylindrical, threaded cylindrical, conical, square, and triangular shapes control the complexity of the material flow and the quality of the weld.

Apart from the tool pin, the tool shoulder design also affects the weld quality. The tool shoulder can be flat, convex, concave, or can have features such as scrolls or grooves. The tool shoulder profile affects the heat generation as well as material flow during welding. Usually, the tool shoulder and pin both rotate during FSW. As the heat requirement in case of joining of PMCs is low, a rotating shoulder can lead to excess heat generation. A stationary shoulder tool is also preferred for the joining of PMC to reduce heat generation. A thrust bearing can be embedded inside the shoulder to prevent shoulder rotation. A brass ring added at the bottom helps to reduce the tool wear (Laieghi *et al.*, 2019a). Such an FSW tool and its schematic illustrations are shown in **Fig. 4**.

Stationary tool shoulders can be made from different materials such as wood, teflon, aluminum (Al), polycarbonate, and brass. **Fig. 5** shows such stationary tools with different shoulder dimensions based on the welding requirements (Eslami *et al.*, 2015).

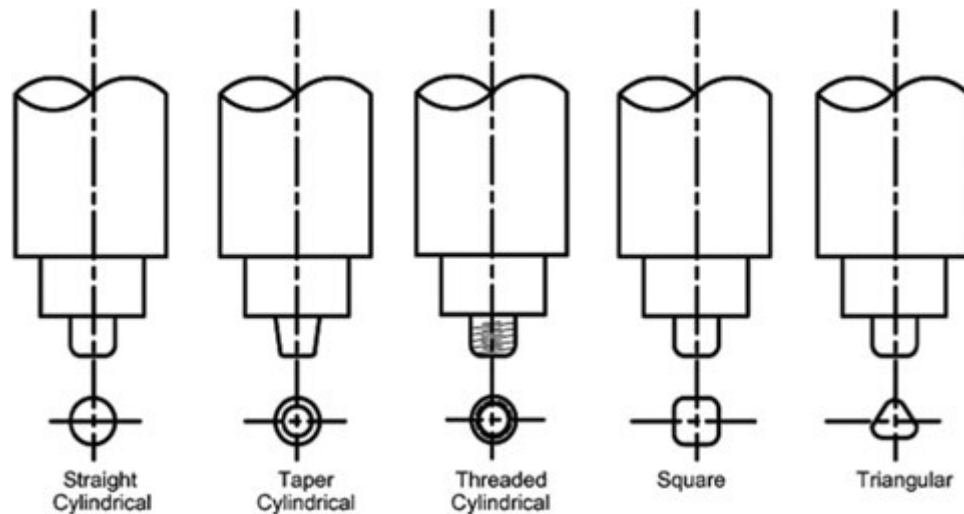


Fig. 3 General tool and pin designs used for FSW. Reproduced from Padmanaban, G., Balasubramanian, V., 2009. Selection of FSW tool pin profile, shoulder diameter and material for joining AZ31B magnesium alloy – An experimental approach. *Materials and Design* 30 (7), 2647–2656.

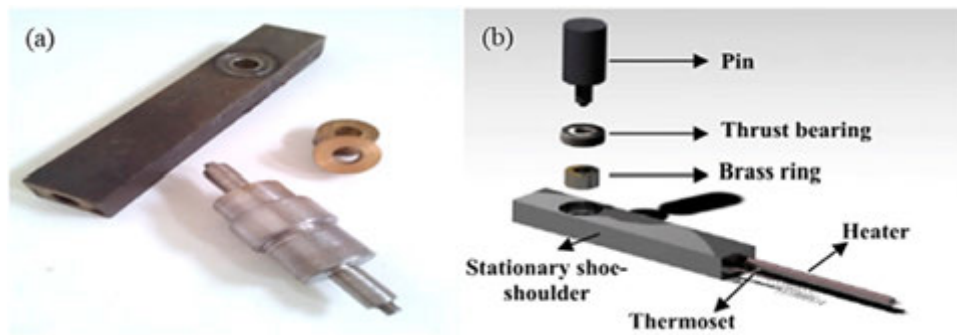


Fig. 4 (a) Actual picture and (b) Schematic illustration of the FSW tool. Reproduced from Laieghi, H., Alipour, S., Mostafapour, A., 2019a. Heat-assisted friction stir welding of polymeric nanocomposite. *Science and Technology of Welding and Joining* 25, 56–65. Available at: <https://doi.org/10.1080/13621718.2019.1610613>.

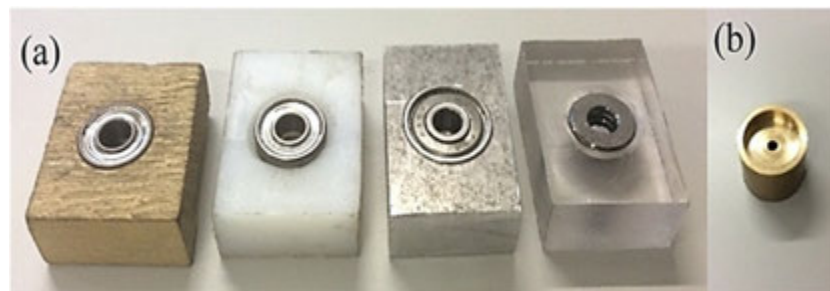


Fig. 5 (a) Stationary shoulders made from wood, teflon, aluminum, and polycarbonate (from left to right) (b) Stationary shoulder made from brass. Reproduced from Eslami, S., *et al.*, 2015. Shoulder design developments for FSW lap joints of dissimilar polymers. *Journal of Manufacturing Processes* 20, 15–23.

A non-heated smoothing shoe can also be used instead of the rotating shoulder (Eslami *et al.*, 2017). Polymers exhibit a poor weld surface quality as compared to metals due to their softer structure and ability to degrade at much lower temperatures (Banjare *et al.*, 2017). A superior weld can be obtained using a static shoulder welding shoe made of PTFE as shown in Fig. 6 (Eslami *et al.*, 2017).

External heating has also been explored for the joining of the polymer-based materials due to the low thermal conductivity of these materials (Banjare *et al.*, 2017). “Hot Shoe” is an effective FSW tool developed with the ability to monitor and control the desired

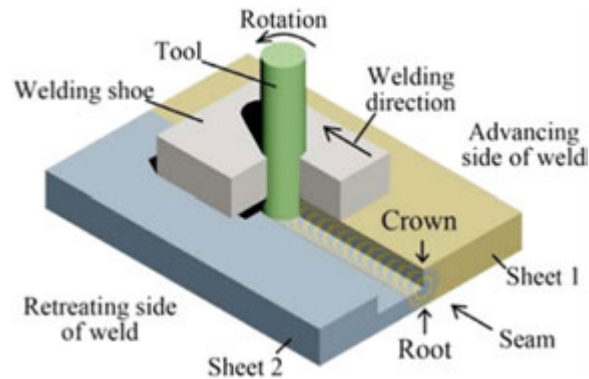


Fig. 6 Schematic of the FSW for polymers with PTFE welding shoe. Reproduced from Eslami, S., Tavares, P.J., Moreira, P.M.G.P., 2017. Friction stir welding tooling for polymers: Review and prospects. *The International Journal of Advanced Manufacturing Technology* 89, 1677–1690.

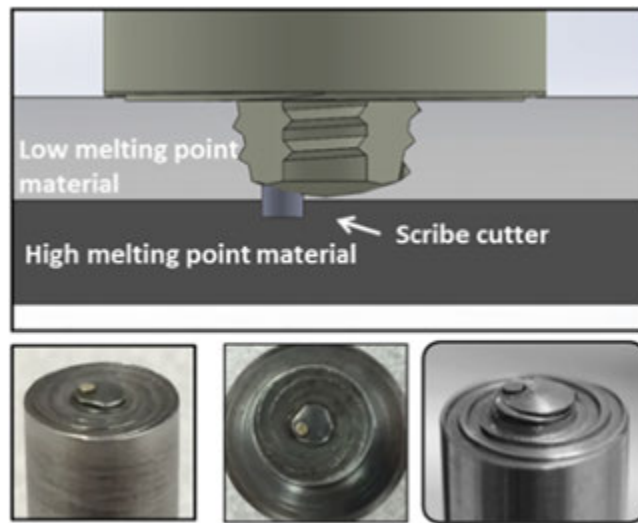


Fig. 7 Schematic of FSS tool as it plunges into a dissimilar material lap joint (above) and different FSS tools (below). Reproduced from Upadhyay, P., *et al.*, 2017. Joining dissimilar materials using friction stir scribe technique. *Journal of Manufacturing Science and Engineering, Transactions of the ASME* 139 (3), 8–10.

amount of heat based on the melting point of the materials to be joined. Joining of materials having different melting points is another major concern in the field of FSW of polymer based materials. This issue can be resolved by modifying the tool design. A scribe cutter is provided at the end of the tool pin. In-situ mechanical interlocks are created between the material interfaces during Friction Stir Scribe (FSS) technique. A typical FSW tool with such a modification enables enhanced material flow in the FSS process. [Fig. 7](#) shows a scribe cutter attached at the tooltip with the offset from the tool rotational axis ([Upadhyay et al., 2017](#)).

Processes and Experimental Procedure

Friction Stir Welding Configuration

Friction stir welding of PMCs can be performed in either butt configuration or the lap configuration. The process parameters during lap and butt welding affect the weld feasibility and weld quality.

Butt welding configuration

Friction stir welding is commonly performed in butt welding configuration where two workpieces are joined by plunging a non-consumable tool at the interface of the workpieces and traversed along the joint direction. Butt-welded polymer joints possess enhanced mechanical properties. The welding is carried out with or without an external heating source. The butt welding configuration can be used for the welding of ex-situ polymer composite materials or the configuration can be used to fabricate

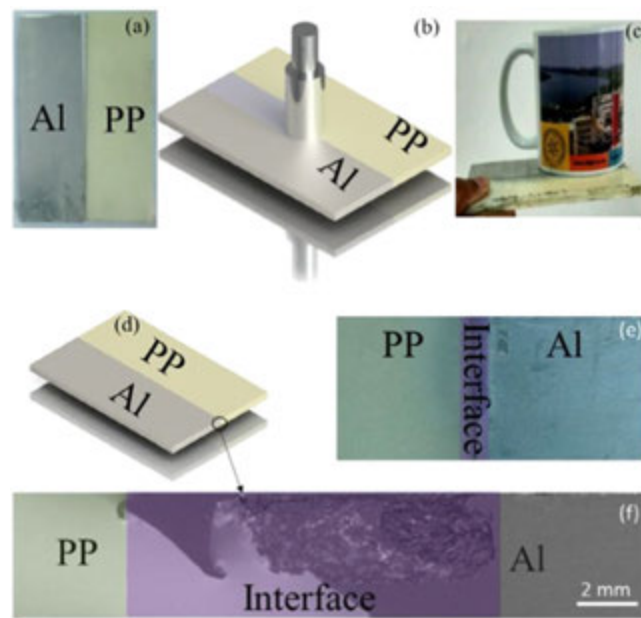


Fig. 8 (a) Aluminum and Polypropylene workpieces before joining (b) Schematic diagram of FSW (c) Al and PP workpieces after FSW (d) Schematic diagram of Al and PP after FSW (e) Top view of dissimilar Al-PP joint (f) Cross-section view of Al and PP weld. Reproduced from Rout, A., *et al.*, 2019. Atomically locked interfaces of metal (aluminum) and polymer (polypropylene) using mechanical friction. *Polymer* 169 (1), 148–153.

in-situ polymer composite materials. The in-situ polymer composites are fabricated by adding reinforcement in the butt weld line during the welding. The reinforcement particles generally used in polymer matrix composite materials are copper (Cu) nanoparticles, Al powder, carbon short fibers, multi-walled carbon nanotubes, glass fiber, basalt fiber, silicon carbide (SiC), alumina (Al_2O_3), graphite, and silica (SiO_2) (Mendes *et al.*, 2014; Rezaee Hajideh *et al.*, 2018; Kumar *et al.*, 2019a,b). Welds with good quality can be achieved in the absence of external heating with optimal tool rotational speed and axial force. Porosity and cavity formation at the Retreating Side (RS) of the stir zone is avoided and a uniform weld region is obtained with optimum process parameters. FSW with a stationary shoulder is used to join acrylonitrile butadiene styrene workpieces without an external heating system (Mendes *et al.*, 2014). Poor mixing of materials at the RS and voids in the nugget region are observed for low rotational speed and smaller axial force due to poor thermal conductivity of polymers. Moreover, in the absence of an external heat source, the heat conduction from the advancing to the retreating side is restricted due to the poor thermal conductivity of ABS. Hence, an external heat source is recommended to have the uniform heat conduction from the advancing to the retreating side. Aluminum (Al) metal and Polypropylene (PP) is welded in the butt welding configuration to enhance the joint efficiency and mechanical properties (Rout *et al.*, 2019). Fig. 8 shows the PP and Al workpieces before and after welding using a butt configuration.

Lap welding configuration

Friction Stir Lap welding (FSLW) is an innovative concept of joining metallic materials, polymers, and polymer matrix composites (Huang *et al.*, 2019). The two workpieces to be welded are placed one over the other and the rotating tool is inserted through one workpiece and is touching the second workpiece. As the two workpieces are one over the other, the tool design is to be carried out wisely to avoid the movement of the workpieces and they would be fastened properly to avoid the weld distortions. Compared to lap configuration, butt configuration can have a general simple tool design. A double-sided bobbin tool is an innovative option that can be used in the lap joint configuration for better welding. Lap configuration welding can be performed with and without an external heating system based on the type of material being welded. The welding of polymer to polymer, metal to polymer, PMC to PMC, and PMC to metals will have different temperature requirements based on which the use of an external heating system can be decided.

The FSLW can also be used for the joining of metal with polymeric materials, such as AA 5058 welded with poly-methyl methacrylate in lap configuration (Derazkola *et al.*, 2018). Another study on the lap joint configuration has reported the successful joining of PMCs with the metal. The joining of a 2060-T8 workpiece with short carbon fiber reinforced poly-ether-ether-ketone (PEEK) is carried out using lap welding (Huang *et al.*, 2019). In the lap joint configuration, the severe plastic deformation occurs at lower peak-temperature compared to the butt-welding configuration. Therefore, better mechanical interlocking is identified in the joining of dissimilar materials.

High shear strength over a wide range of welding parameters is exhibited by the FSLW of aluminum alloy AA 6061 and monomer casting (MC) Nylon-6 (Liu *et al.*, 2014). FSLW is also used to prepare a hybrid joint using a sufficient mixture of aluminum 6061-T6 alloy and PEEK using a tapered thread pin with the triple facets (Huang *et al.*, 2018a). FSLW result in higher joint efficiency for the dissimilar joining of Poly Lactic Acid (PLA) material and PLA reinforced with 30 wt% basalt fiber. The higher joint efficiency is a result of the broken long basalt fibers into smaller pieces, which could not go over the barrier between the base

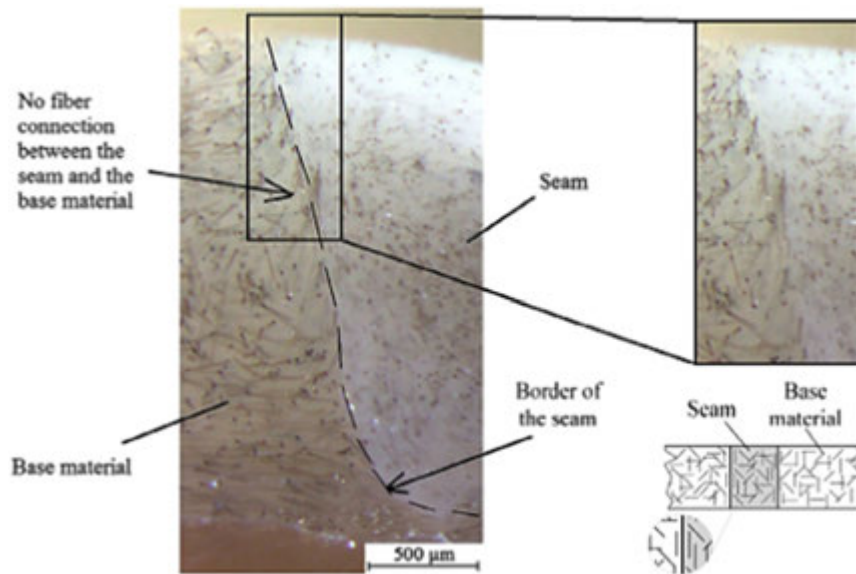


Fig. 9 FSW of a thin sheet made of PLA with 30% basalt fibers reinforcement. Reproduced from Kiss, Z., Temesi, T., Czigány, T., 2018. Adherability and weldability of poly(lactic acid) and basalt fibre-reinforced poly(lactic acid). *Journal of Adhesion Science and Technology* 32, 173–184.

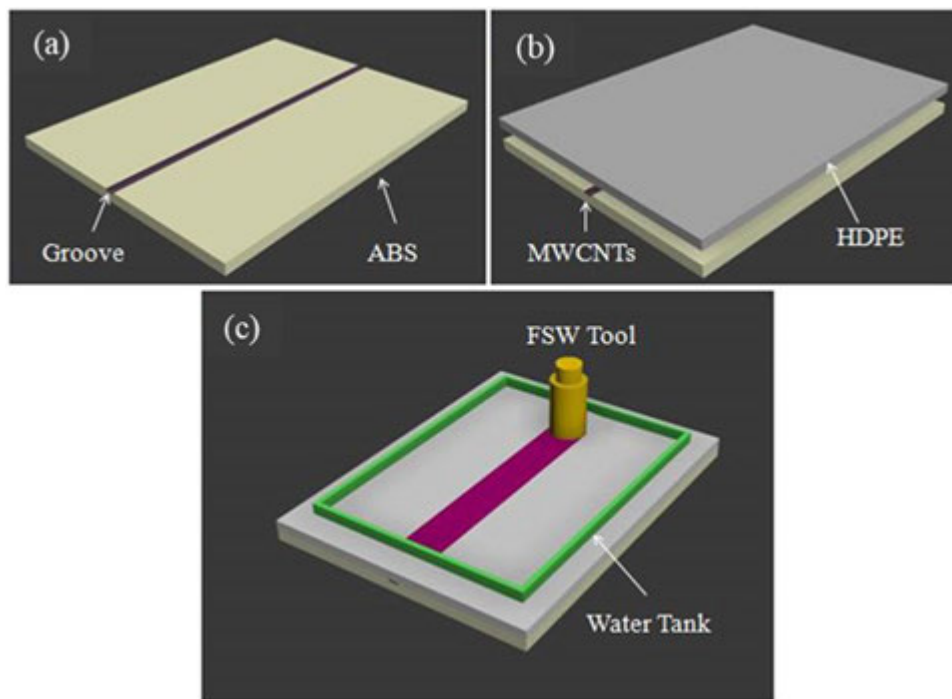


Fig. 10 Schematic of the submerged FSW on dissimilar thermoplastics joint. Reproduced from Gao, J., *et al.*, 2015. Improvements of mechanical properties in dissimilar joints of HDPE and ABS via carbon nanotubes during friction stir welding process. *Materials and Design* 86, 289–296.

material and the weld. The long basalt fibers in the base material are broken down into shorter lengths in the seam area as shown in Fig. 9 (Kiss *et al.*, 2018).

Submerged FSW

Submerged FSW is an extended application of FSW where the workpiece is immersed in a liquid medium to overcome the overheating issues during the welding of polymers and PMCs. A stationary liquid bath or flowing water supply can be used for the submerged FSW. Fig. 10 shows the schematic of lap of HDPE and ABS embedded with the multi-walled carbon nanotube.

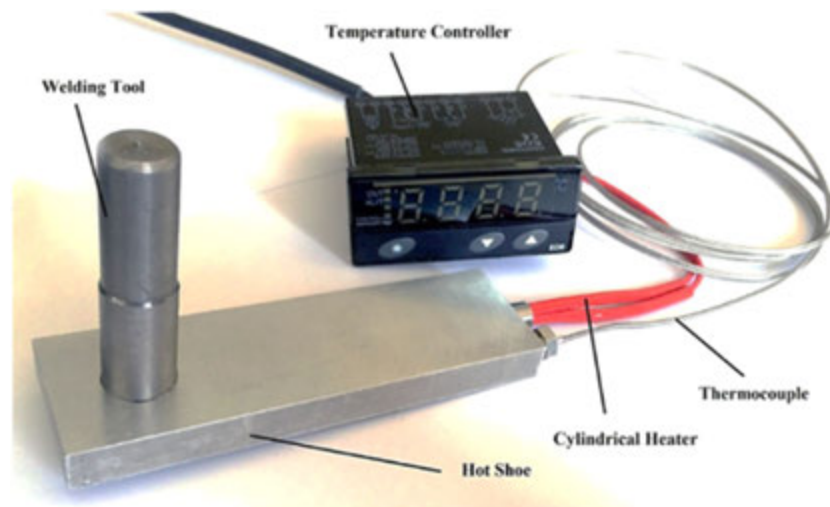


Fig. 11 Additional heating set-up for the friction stir welding of reinforced dissimilar polymer friction stir welding. Reproduced from Rezaee Hajideh, M., Farahani, M., Molla Ramezani, N., 2018. Reinforced dissimilar friction stir weld of polypropylene to acrylonitrile butadiene styrene with copper nanopowder. *Journal of Manufacturing Processes* 32, 445–454.

A groove is cut in the longitudinal direction in the ABS workpiece. Multi-walled carbon nanotubes are filled and compressed in the groove. The HDPE is placed covering the ABS workpiece as shown in Fig. 10(b). A water tank is kept on the HDPE to carry out submerged FSW (Gao *et al.*, 2015).

Heat-Assisted FSW

An external heating system can be incorporated during the FSW of polymer based materials to enhance the weld quality and reduce the defects. The cooling rate during the welding affects the weld quality and the heat-assisted FSW is a procedure to avoid the thermal defects which arise during welding. The external heating system helps in the mixing and joining of polymeric materials during the welding to achieve successful welds with superior properties (Rezaee Hajideh *et al.*, 2018). One such external heating system is shown in Fig. 11.

The polymer sheets welded using heat-assisted FSW exhibit enhanced tensile strength. Heat assisted FSW also achieved reduced chip generation, good surface finish of weld, and minimal material wastage during the welding of polypropylene workpieces (Banjare *et al.*, 2017). Also, the heat-assisted FSW workpieces showed better tensile strength and ductility compared to the polymers welded without a heating system. The heat-assisted FSW is a solution to the welding issues such as lack of proper fusion between the polymeric materials, reduced amount of heat, and generation of voids and porosities.

The feasibility of FSW of polyamide 6/Nitrile butadiene rubber composite is carried out with an additional heating system. The sound welds are obtained by controlling heat input and cooling rate during heat-assisted FSW (Derazkola *et al.*, 2018). The influence of heat-assisted FSW processing parameters and the amount of halloysite nanotubes (HNT) reinforcement on the composite mechanical properties are studied using response surface methodology. Tool rotational speed of 900 rpm, traverse speed of 14 mm/min, and 6.72% HNT are found to be the optimized parameters using the Analysis of Variance Method. The most effective variable among these three is the tool rotational speed followed by the amount of HNTs and the tool traverse speed (Laieghi *et al.*, 2019b).

Friction Stir Spot Welding

Friction stir spot welding (FSSW) is a variant of the friction stir welding process where only plunge and tool retraction is used to spot-weld the workpiece material. Therefore, FSSW can be used to replace resistance spot welding, riveting, clinching, or any similar single point joining processes. FSSW is used to weld the polymeric materials such as High-Density Poly-Ethylene (HDPE), Poly-Propylene (PP), Poly-Methyl-Meth-Acrylate (PMMA), and Acrylonitrile Butadiene Styrene (ABS) sheets.

The processing of FSSW is much similar to FSW. The different stages of the FSSW schematic are illustrated in Fig. 12 (Lambiase *et al.*, 2015). The tool is rotated at the prescribed speed and moved down towards the stationary workpiece. As the rotating tool is plunged in the upper sheet of the workpiece, frictional heat is generated near the tool pin interface. Further, when the tool is plunged in the sheets at a constant speed, a certain part of the material is ejected from the workpiece as shown in Fig. 12. The plunging process is continued until the shoulder touches the workpiece surface, whilst the tool rotation is still consolidated for the dwell time. The material underlying and surrounding the tool is heated and softened. The tool shoulder compresses the softened material. The tool rotation is stopped and the tool is made stationary before retraction. Cross-section image of friction stir spot welded white and transparent polycarbonate sheets shown in Fig. 13 exhibit the material mixing and the ejected material during the welding.

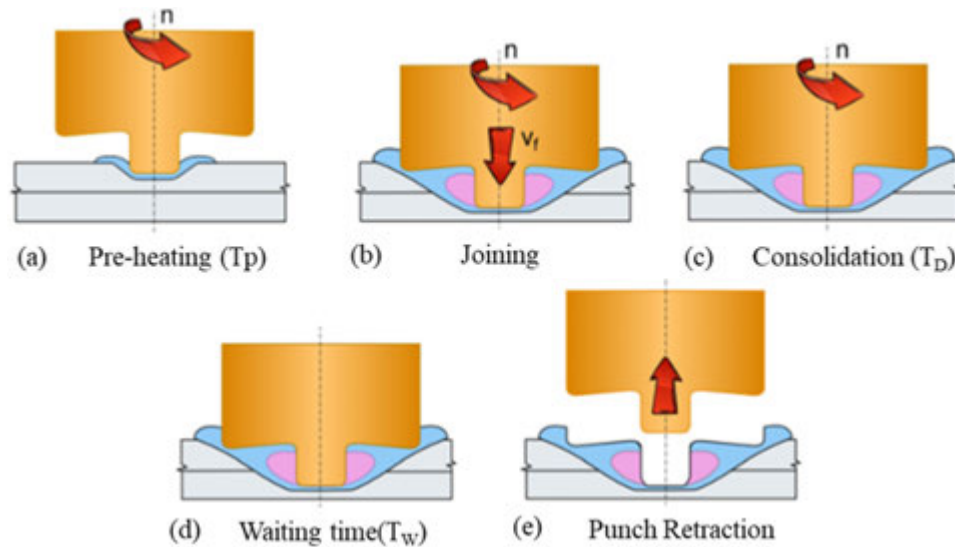


Fig. 12 Different stages of friction stir spot welding. Reproduced from Lambiase, F., Paoletti, A., Di Ilio, A., 2015. Mechanical behaviour of friction stir spot welds of polycarbonate sheets. *International Journal of Advanced Manufacturing Technology* 80 (1–4), 301–314.

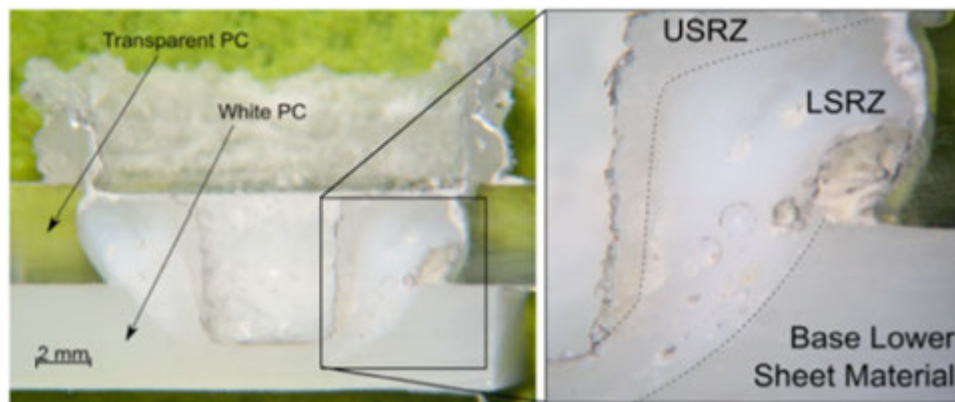


Fig. 13 Cross-section of friction stir spot welded polycarbonate sheets. Reproduced from Lambiase, F., Paoletti, A., Di Ilio, A., 2015. Mechanical behaviour of friction stir spot welds of polycarbonate sheets. *International Journal of Advanced Manufacturing Technology* 80 (1–4), 301–314.

Friction stir spot welding is also used to join Poly-Ether-Imide (PEI) laminates reinforced with carbon fiber and the fracture mechanism of these joints has been studied. Rigidity, high strength, chemical resistance, and low water absorption have made carbon fiber reinforced PEI (CF-PEI) laminate a suitable candidate material for aerospace parts. The carbon fibers lead to the macromolecular inter-diffusion and interlocking at the interfaces leading to strong bonding between the Sleeve Stirring Zone (SSZ) and Thermo-Mechanically Affected Zone (TMAZ) (Huang *et al.*, 2018c).

Friction Riveting

Friction riveting is considered as an intermediate technique of mechanical fastening and welding techniques. Friction riveting offers certain advantages such as short joining cycles and fewer surfaces pre-treatment. The installation steps are reduced as pre-drilling is not required for the friction riveting. The main processing steps of friction riveting are friction step, forging step, and consolidation illustrated in Fig. 14(a) (Altmeyer *et al.*, 2015). The rotating rivet is plunged into the polymer workpiece. At the same time, friction and axial pressure generate the heat. The polymer layer which is in contact with the tip is melted due to the frictional heat generation. This material is expelled partially and forms a flash under the effect of continuous axial loading at an increased temperature. The rivet tip is also plasticized due to temperature rise. The axial force is further increased and spindle rotation is decelerated using a motor brake. The molten polymer and the extruded rivet tip are suppressed by the axial loading. Finally, the riveted joint is consolidated and cooled down at a constant external pressure. The variation in processing parameters during the different stages of friction riveting is illustrated in Fig. 14(b) (Altmeyer *et al.*, 2015).

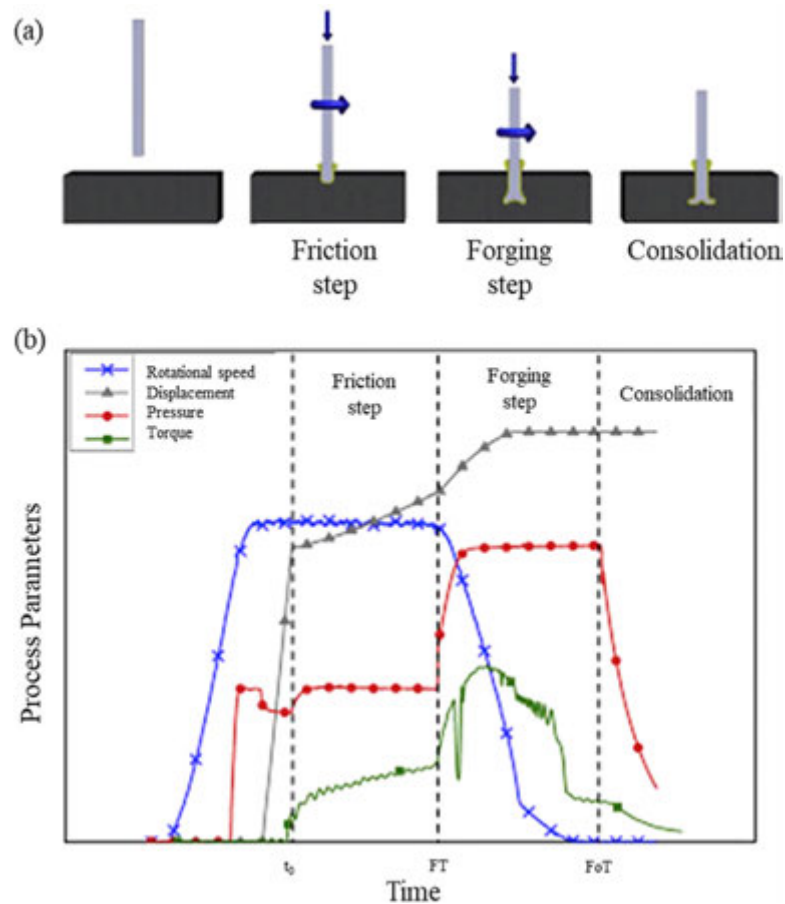


Fig. 14 Friction riveting process: (a) Process steps and (b) Process parameter evolution over time. Reproduced from Altmeyer, J., *et al.*, 2015. Microstructure and mechanical performance of metal-composite hybrid joints produced by FricRiveting. Composites Part B: Engineering 81, 130–140.

Friction Stir Processing

Friction stir processing (FSP) is derived from the basic principles of FSW. FSP is generally used for surface microstructural modification and surface composite fabrication. FSP is also a solid-state method used for the PMC fabrication. The PMC materials fabricated using FSP exhibit enhanced material properties. The PMCs can be fabricated using metallic, ceramic, or polymer reinforcement. The addition of nano-clay particles into Ethylene-Propylene Diene Monomer (EPDM) polymer matrix enhanced tensile strength by $\sim 15\%$ (Nakhaei *et al.*, 2016). Polymer particles can also be added as reinforcement to the metal matrix to fabricate metal matrix composite materials. FSP is used to fabricate Al matrix composites by the addition of polymer particles such as a low-density polymer (polyethylene terephthalate) to enhance mechanical and electrochemical properties of the base Al alloy (Rout *et al.*, 2020). FSP is also used to fabricate metallic and ceramic particle reinforced metal matrix composites with enhanced mechanical and electrochemical properties (Mahesh and Arora, 2019; Mahesh *et al.*, 2020).

Microstructure Evolution and Material Flow Analysis

The properties of the FSW components are majorly dependent on the microstructural features and the material flow during the welding. Hence, understanding the microstructure and the material flow during FSW is vital. The heat generated during the welding process influences the material flow. The decision on the use of an external heating arrangement can be made based on the material flow analysis. A heat-assisted FSW can be used if a higher processing temperature is required. Heat generation and severe plastic deformation during FSW result in the formation of different microstructural zones in the weld region. The different regions during FSW of metallic materials shown in Fig. 15 are identified based on the microstructural variations. The major classifications are stir-zone or nugget-zone (SZ or NZ), TMAZ, and HAZ (Nimer *et al.*, 2013).

FSW is also helpful in reducing the size of embedded phases in NZ. The average size of the base spherulitic phase is reduced to half after the welding of polypropylene polymer workpiece. A transition zone is observed at the border of the weld zone and base material. The width of the transition zone is governed by FSW parameters such as tool rotational speed and tool traverse speed.

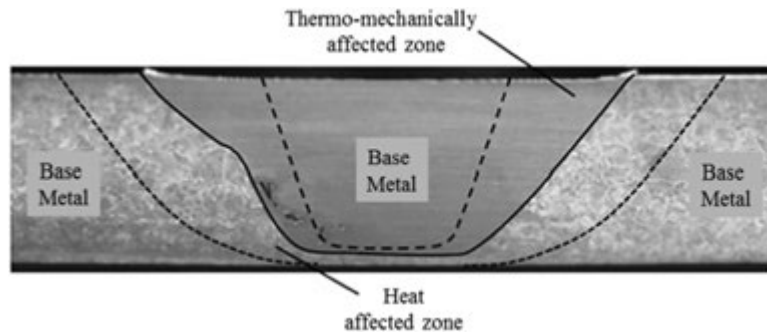


Fig. 15 Friction stir weld cross-section with different regions. Reproduced from Nimer, S., Wolk, J., Zupan, M., 2013. Local property characterization of friction stir welded Ti-5111: Transverse orientation measurements. *Acta Materialia* 61 (8), 3050–3059.

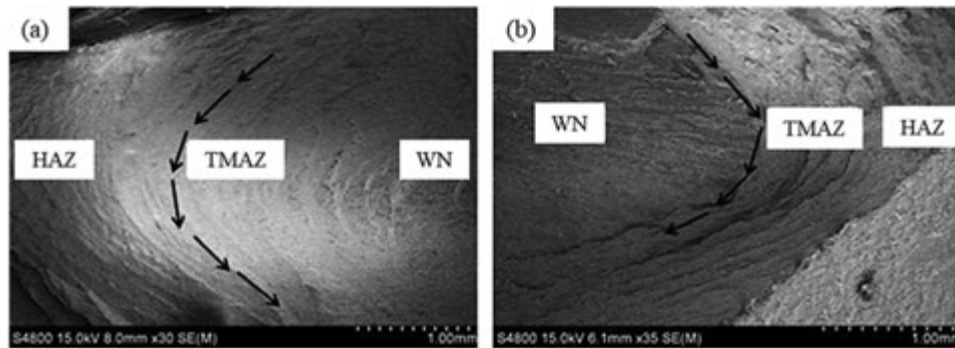


Fig. 16 SEM micrographs of the friction stir welded HDPE and ABS with carbon nanotubes (a) AS (b) RS. Reproduced from Gao, J., *et al.*, 2015. Improvements of mechanical properties in dissimilar joints of HDPE and ABS via carbon nanotubes during friction stir welding process. *Materials and Design* 86, 289–296.

HAZ is influenced by the heat generation only. The identification of such phases in the weld sections can be carried out using SEM as shown in **Fig. 16**. Another important feature is the ring formation in the polymer matrix due to the material flow during the welding. The ring shape is not symmetric in AS and RS owing to the different material flow (Gao *et al.*, 2015).

The microstructural features such as NZ, HAZ, and TMAZ are visible in butt welding configuration whereas the lap welding configuration microstructural features are different when a metallic plate is welded with a polymer workpiece. The softer polymer will mix into the metallic base plate and the interlocking between the polymer and metallic phase creates the weld. The generated heat is insufficient to melt the metal and an interlock in the polymer matrix with metal is created due to the thermo-mechanical deformation.

Material flow is crucial to analyze basic features of friction stir welds. It helps understand defect formation and ways to reduce such defects. The material flow is studied using different techniques for metallic materials such as X-ray computed tomography, use of transparent or dissimilar materials, markers, broken pin, and keyhole formation (Pandya *et al.*, 2019). Frictional heat generation and material flow result in the formation of the weld zone. Excessive heat generation leads to partial melting of polymer workpiece and thereby generates poor weld quality (Huang *et al.*, 2019). For example, when the welding is carried out between a metal and a polymer, welding defects are generated due to different thermal behaviors. The intermixing of PMMA and AA 5058 in the stir zone during the FSW is shown in **Fig. 17** (Derazkola *et al.*, 2018). The material in the vicinity of the tool is deformed with the tool rotation due to severe plastic deformation. The heat is not sufficient enough to melt the aluminum alloy and creates an interlock in the polymer matrix under the thermo-mechanical deformation. A very thin TMAZ and significant HAZ are observed in aluminum alloys. The material flow in advancing and retreating sides is not uniform due to different temperature distribution on both sides. Lower heat generation at the retreating side form higher fragments. A higher circular flow of fragmented aluminum at the advancing side indicates an enhanced flow of aluminum in the molten polymer matrix. The small fragments at the advancing side are attributed to the severe plastic deformation at a higher peak temperature. A layered structure of polymer and metal matrix is identified at the center of the U-shaped weld zone. The SEM images of material flow in AS, center, and RS are shown in **Fig. 17(d)–(f)**.

The material flow during welding can be turbulent based on the temperature generated during welding. A mixed interface shows a turbulent material flow on the AS of the aluminum-polymer interface as shown in **Fig. 18**. However, a separate interface is observed at RS which shows the absence of a turbulent flow. Semi-crystalline phases are formed at the aluminum-polymer interface due to the intermixing of aluminum and polymer at a higher temperature. Such a semi-crystalline phase in the thickness range of 20 nm identified using Transmission Electron Microscopy (TEM) is shown in **Fig. 18(c)** (Derazkola *et al.*, 2018).

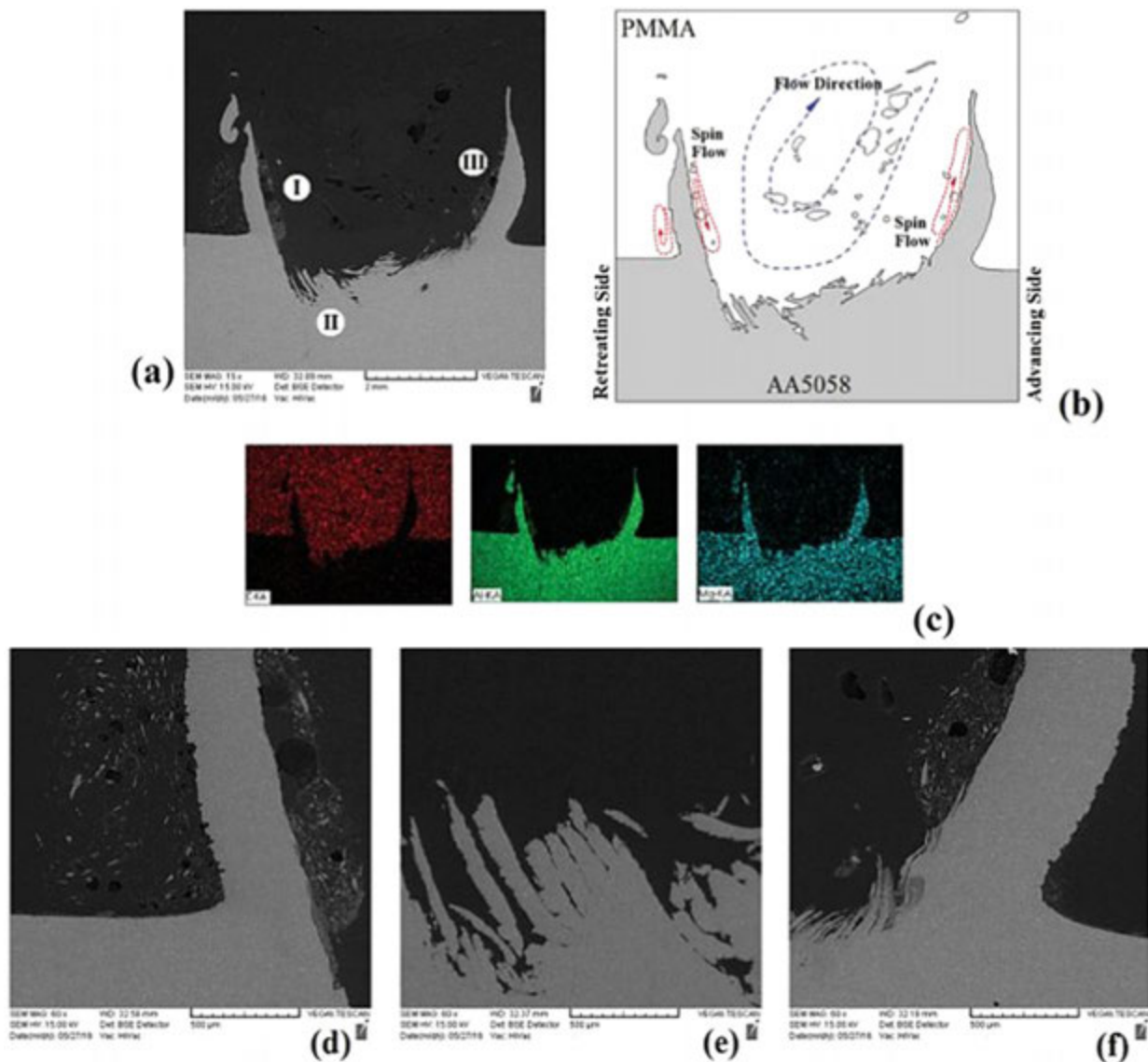


Fig. 17 Cross-sectional view of the dissimilar joint perpendicular to FSW direction: (a) SEM image, (b) schematic representation of materials flow pattern, and (c) elemental mapping chemical analysis. SEM images from (d) RS (e) center and (f) AS. Reproduced from Derazkola, H.A., Khodabakhshi, F., Simchi, A., 2018. Friction-stir lap-joining of aluminium-magnesium/poly-methyl-methacrylate hybrid structures: Thermo-mechanical modelling and experimental feasibility study. *Science and Technology of Welding and Joining* 23, 35–49.

The tool rotational speed plays a crucial role in the material flow during the dissimilar metallic-polymer lap welding of AA 2060-T8 and SCF/PEEK. The anchor feature developed during welding at high tool rotational speeds enable strong interlocking between the metallic and polymeric workpieces as shown in Fig. 19. The aluminum alloy AA 2060-T8 is stirred along with the tool rotation and gets inserted in the polymer matrix. The anchors form a strong interlocking at the AA 2060-T8 and SCF/PEEK interface. Large size anchors form at low tool rotation speed as the stirring ability is not sufficient enough to break these anchors. At the higher rotational speed, these anchors are broken due to significant stirring action. Hence, the height and width of these anchors are reduced as shown in Fig. 19(b). In the case of very high rotational speed, the material is softened to cause aluminum anchors to deform and bend as shown in Fig. 19(d) (Huang *et al.*, 2019).

Processing Parameters

Most of the welding defects during FSW can be minimized by the use of appropriate processing parameters and the welding tool designs. The defects such as flash and tunnel defects can be controlled by the optimization of processing parameters. One

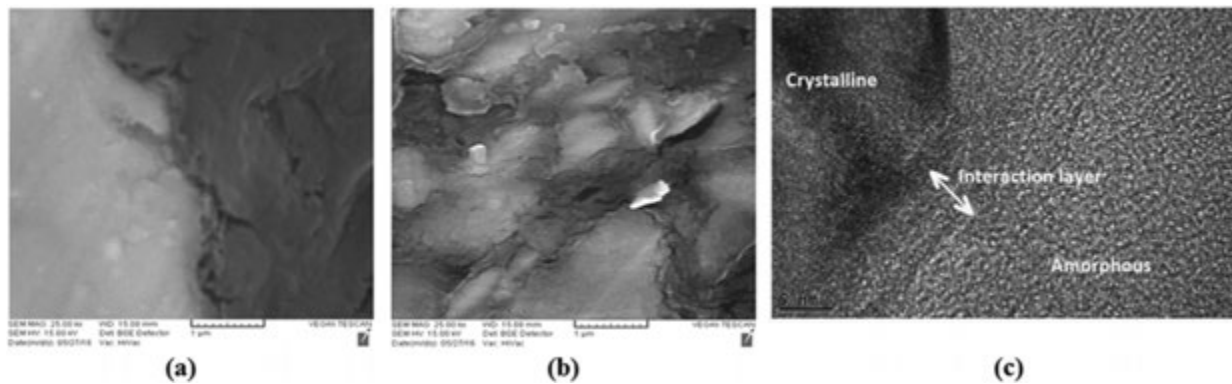


Fig. 18 High magnification SEM images of Al-polymer interface (a) RS and (b) AS, (c) HR-TEM image showing the atomic structure at the interactional bonding layer. Reproduced from Derazkola, H.A., Khodabakhshi, F., Simchi, A., 2018. Friction-stir lap-joining of aluminium-magnesium/poly-methyl-methacrylate hybrid structures: Thermo-mechanical modelling and experimental feasibility study. *Science and Technology of Welding and Joining* 23, 35–49.

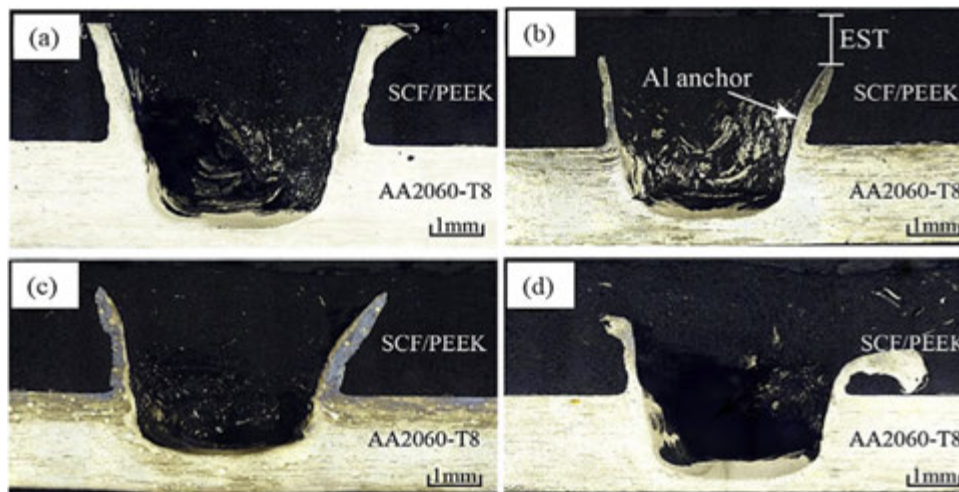


Fig. 19 AA 2060-T8 and SCF/PEEK friction lap weld interface macrostructures at (a) 1400 rpm, (b) 1600 rpm, (c) 1800 rpm and (d) 2000 rpm. Reproduced from Huang, Y., *et al.*, 2019. Improving mechanical properties of composite/metal friction stir lap welding joints via a taper-screwed pin with triple facets. *Journal of Materials Processing Technology* 268 (October), 80–86.

parameter used in the study on the effectiveness of friction stir welded materials is K-factor (Kiss and Cziga, 2012). K-factor is defined based on the three main process parameters viz., tool rotational speed (ω), feed rate (v), and tool diameter (D).

$$K - \text{factor} = \frac{\omega}{v} \cdot D$$

The FSW of polyethylene-terephthalate-glycol fabricated using various welding tools and parameters are analyzed by correlating the K-factor which is related to the welding heat input. The K-Factor is found to be in direct correlation with the HAZ range and the mechanical properties of the fabricated FSW seams. The feeding rate and the heat transfer are inversely proportional and tool rotational speed and tool diameter are directly proportional to the heat transfer. Table 1 shows the K-factor for different welding parameters (Kiss and Cziga, 2012).

The weld quality is decreased if K-factor < 150 as the softening of the materials to be welded does not take place and some non-melted particles are observed on the welds which might even lead to the welding tool breakage. On the contrary, a very high temperature is generated when the K-factor is too high (K-factor > 500). The material is melted completely and is pushed out at this temperature. As a result, the defective weld is generated for a very high K-factor value. Hence, a perfect weld is obtained for a certain range of K-factor values.

Another parameter used to analyze the weld quality is a linear relationship between the welding parameters such as tool rotational speed (ω) and the welding speed (v). As shown in Fig. 20(a) the thickness increases with an increase in tool rotational speed for a given tool traverse speed. The thickness decreases with increasing welding speed. The thermal input is increased and the Al 6061 workpieces are bent towards the Nylon-6 side with an increase in the rotation rate. The bubble volume in the joints is reduced and the nominal shear stresses are increased at the joints due to an increase in the tool rotation rate. The effect of a

Table 1 K-Factor with welding parameters and tools

Rotational speed (rpm)	Feed (mm/min)	Tool diameter (mm)	K-Factor
750	75	8	80
900	100	8	72
1200	50	8	192
1200	90	12	160
1200	50	12	288
1200	100	8	96
2550	100	8	204
2550	50	8	408

Note: Kiss, Z., Cziga, T., 2012. Effect of welding parameters on the heat affected zone and the mechanical properties of friction stir welded. Journal of Applied Polymer Science 125, 2231–2238.

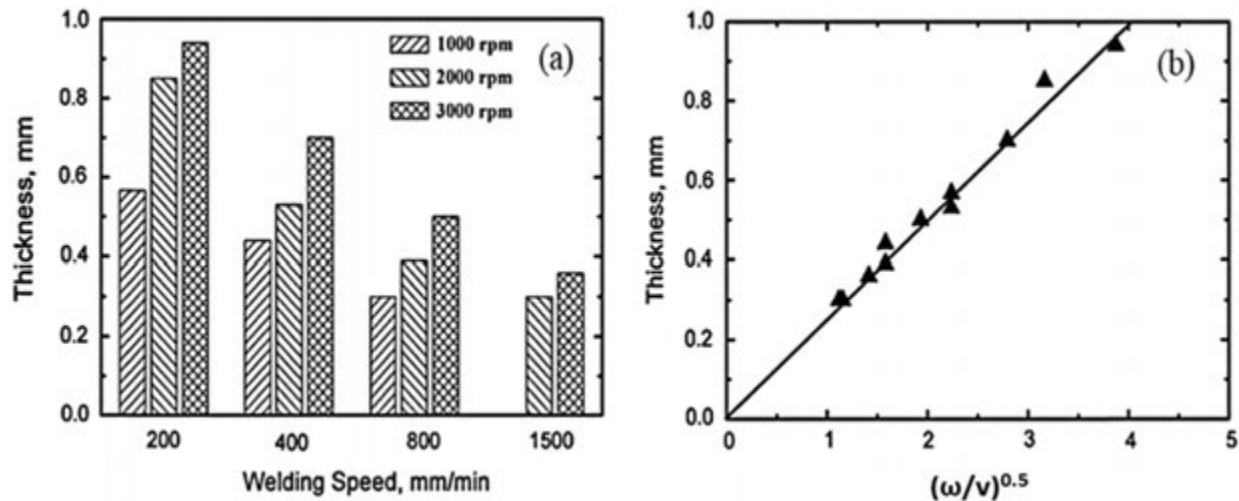


Fig. 20 Variation in thickness of melted nylon with (a) various welding speeds and rotational speed, and (b) $(\omega/v)^{0.5}$. Reproduced from Liu, F.C., Liao, J., Nakata, K., 2014. Joining of metal to plastic using friction lap welding. Materials and Design 54, 236–244.

parameter $(\omega/v)^{0.5}$ and thickness (H) of melted nylon are used to understand the quality of welding of aluminum alloy Al 6061 and MC Nylon-6 as shown in Fig. 20(b) (Liu *et al.*, 2014).

Tool Rotational Speed

Lower tool rotational speed leads to poor heat generation. However, higher tool rotational speed might lead to excessive heat generation which might disintegrate the polymer chains. Polymer degradation and melting occur due to this excessive heat generation. The mechanical properties also deteriorate at higher tool rotational speed (Huang *et al.*, 2018b; Laieghi *et al.*, 2019a). The variation in the material properties based on the change in the tool rotational speeds such as 1400, 1600, 1800, and 2000 rpm is shown in Fig. 21.

The amount of flash generated on the advancing side is found to increase with an increase in the tool rotational speed. The weldment surface is smooth at the higher rotational speed due to better material flow. In the lap joint configuration, the amount of intermixing of the materials is increased with an increase in tool rotational speed (Derazkola *et al.*, 2018).

Tool Traverse Speed

Tool traverse speed defines the time required to complete the welding operation. When the tool traverse speed is too low, over stirring will take place due to longer processing time. Therefore, polymer might degrade due to more heat generation at the lower traverse speed. When the tool traverse speed is too high, the time might be insufficient to fill the weld zone. Further, the defects such as porosities and cavities might be formed due to the insufficient heat generation at higher tool traverse speed (Laieghi *et al.*, 2019a). For a sound weld, the traverse speed should be optimum so that the amount of heat generation is sufficient to effectively pre-heat the material (Huang *et al.*, 2019). AA 6061-T6 aluminum alloy and Polyether Ether Ketone (PEEK) are joined using Friction Stir Lap Welding (FSLW). When the welding speed is increased, the size of Al-anchor is reduced and reduction in the adhesive area is observed which eventually deteriorated the mechanical interlocking (Huang *et al.*, 2018a).

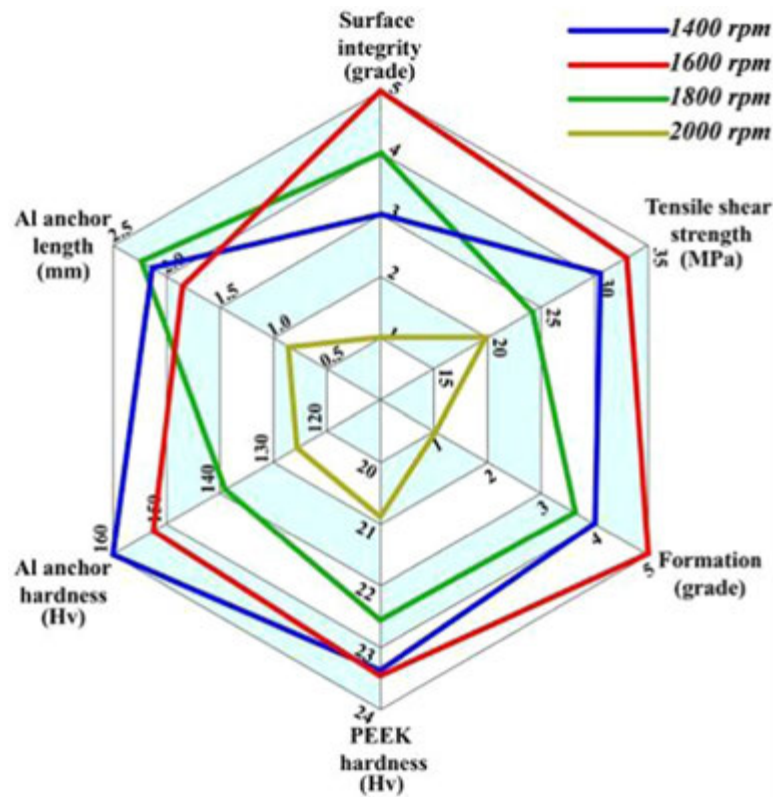


Fig. 21 Schematic representation of the change in different material properties with the tool rotational speeds. Reproduced from Huang, Y., Meng, X., Xie, Y., Li, J., *et al.*, 2018b. Joining of carbon fiber reinforced thermoplastic and metal via friction stir welding with co-controlling shape and performance. *Composites Part A: Applied Science and Manufacturing* 112, 328–336.

Lower weld speeds generate a defect throughout the weld surface due to excessive heat generation. At lower temperatures, the processing width remains uniform. Clear striations are observed at higher weld speeds. This is due to the higher melting and solidification of the material. Depressions along the weld line as shown in Fig. 22 indicate the collapse of the material in the weld zone due to material loss either by ejection or by shrinkage. The tendency to form grooves is decreased with the increase in the tool rotation speed (Sheikh-Ahmad *et al.*, 2019).

Tool Tilt Angle

The angle of the tool center-line with the normal to the workpiece defines the tool tilt angle, with zero tilt angle being the workpiece normal. Different researchers studied the effect of the tool tilt angle (from 0° to 4°) on the weld properties (Zhang *et al.*, 2018). Tilt angle is provided to increase material flow and forging force. However, excessive weld flash generates at the RS for the higher tool tilt angle. Therefore, voids or tunnel defects occur due to the insufficient material to fill the weld cavity. Mechanical properties increase with an increase in tool tilt angle for the metallic materials.

Friction stir welding parameters affect the mechanical properties and weld quality of polymer material welds. Among the welding parameters, tool rotational speed is one important processing parameter which governs the quality and is followed by the traverse speed and tilt angle. The effect of these processing parameters is summarized in Fig. 23. The contribution of tool rotational speed, traverse speed, and tilt angle for the efficient weld joint is identified as 73.85%, 20.18%, and 5.96% respectively (Bozkurt, 2012).

Tool Geometry

Material flow is also influenced by the tool pin geometry. The pin can be cylindrical, conical, triangular, threaded, or cylindrical-conical. The material flow is different as different tool pin features generate different amounts of heat energy. Mechanical interlocking at the weld interface is improved using a taper-threaded tool pin having triple facets. Further, the tilt angle produced sufficient forging force to improve the joint formation (Huang *et al.*, 2019).

The weld region produced during the FSW of polymers using a rotating shoulder does not have a good surface finish. The root defects also are formed along the weld line. Therefore, the rotating shoulder is not considered a feasible option for the friction stir welding of polymers. A stationary shoulder pushes a material inside the weld region and does not allow the softer material to

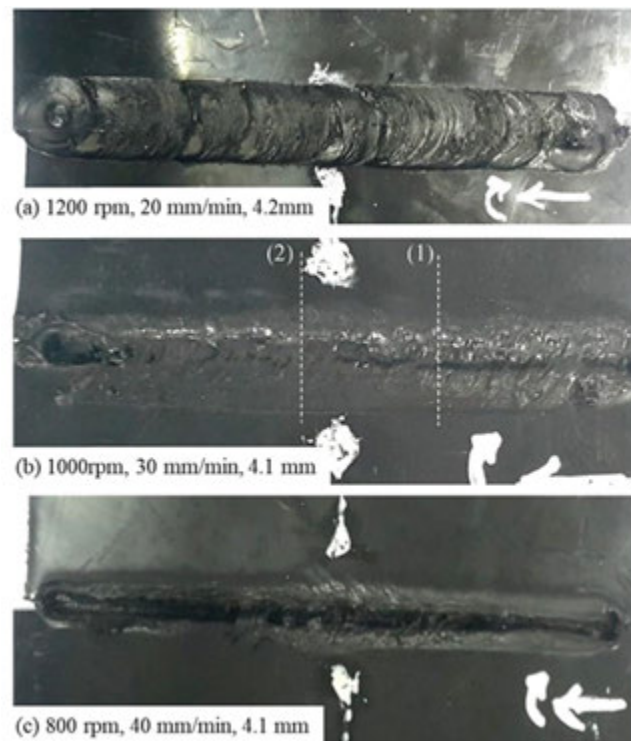


Fig. 22 Weld line appearances at different weld conditions (a) uniform with striations, (b) uniform with surface melting, and (c) groove. Reproduced from Sheikh-Ahmad, J.Y., *et al.*, 2019. Friction stir welding of high density polyethylene – Carbon black composite. *Journal of Materials Processing Technology* 264 (4), 402–413.

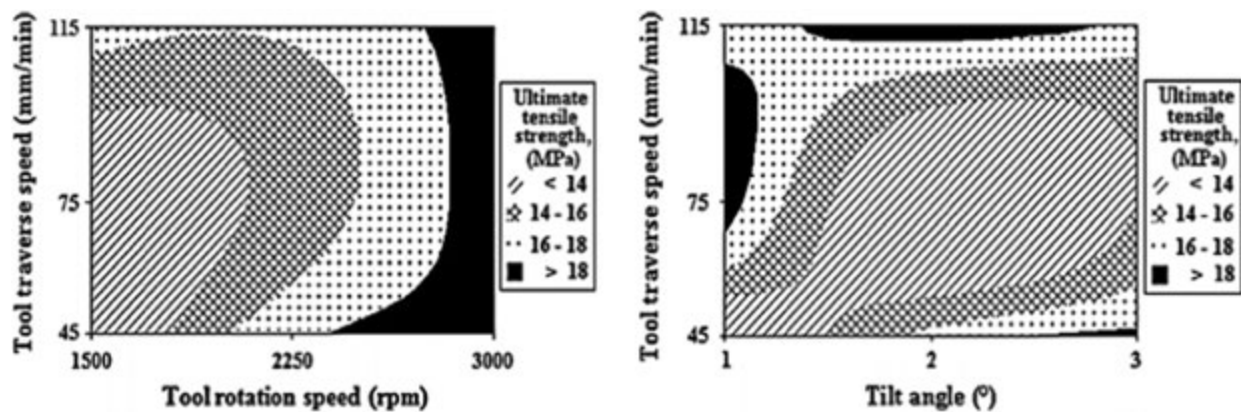


Fig. 23 Optimum tool parameters for the high strength polypropylene weld joints. Reproduced from Bozkurt, Y., 2012. The optimization of friction stir welding process parameters to achieve maximum tensile strength in polyethylene sheets. *Materials and Design* 35, 440–445.

come out. Hence, flash defects can be prevented using a stationary tool shoulder. Different materials can be used to fabricate stationary shoulder for the successful FSW. Better surface quality is observed for the Teflon and polycarbonate stationary shoulders and followed by aluminum and wood, respectively. The thermal conductivity of the brass shoulder is significantly higher compared to the polymer. Therefore, a greater amount of generated heat is transferred through the shoulder and the weld region suffers insufficient heat generation. During plunging, the axial force required for the stationary shoulder is more when the shoulder is engaged with the workpiece. It further reduces as the tool traverses. The tensile strength for the welds obtained using the stationary shoulder is found to be 40% more than the same when rotating shoulder is used. Less amount of heat is generated at the retreating side due to the very low thermal conductivity of polymers. Hence, the specimens fail on the retreating side predominantly due to the lack of stirring effect (Rezaee Hajideh *et al.*, 2018).

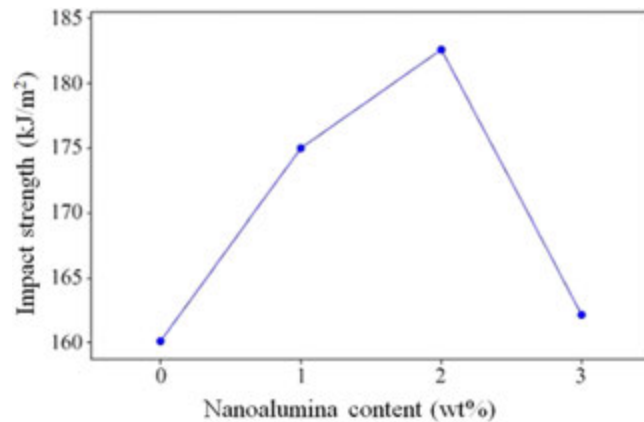


Fig. 24 Effect of nanoalumina content on impact strength. Reproduced from Azdast, T., Hasanzadeh, R., Moradian, M., 2018. Improving impact strength in FSW of polymeric nanocomposites using stepwise tool design. *Materials and Manufacturing Processes* 33 (3), 343–349.

Effect of Reinforcement

Reinforcements in PMCs may be metals, ceramics, or polymers. The rotational movement of the material distributes the reinforcements in the weld regime. The reinforcements such as carbon nanotubes and carbide nano-powders are used to improve the mechanical properties of the polymer. Better weld properties are identified up to a certain amount of reinforcement addition. Mechanical properties start to deteriorate when the amount of reinforcement is more than a certain limit. The higher volume fraction of reinforcement might provide a site for crack initiation and propagation due to agglomeration. These agglomerated particles act as a defect which reduces the mechanical properties of the joint (Gao *et al.*, 2015). The amount of reinforcement is considered to be the most important parameter in the FSW of polycarbonate workpieces where nano-alumina is used as the reinforcement. The nano-alumina reinforcement of 2% significantly increased the impact strength. Increasing the amount of alumina above 2% inversely affects the impact strength due to the agglomeration of alumina particles as shown in Fig. 24 (Azdast *et al.*, 2018).

The fragmentation of reinforcement is another major concern when fibers or short fibers are used as reinforcement. The fibers are mechanically sheared due to the severe plastic deformation of the material during FSW. Weld strength is influenced by the degree of fragmentation of reinforcement fibers in the weld region (Huang *et al.*, 2018c). The fiber disintegration is higher due to shearing forces during tool rotation. This fragmentation of fibers can be identified by the relative frequency analysis.

Mechanical Properties

Mechanical properties of the friction stir welded materials are related to various process parameters and machine variables. As discussed in Section “Processing Parameters”, the properties can be related to the K-factor which is a correlation between tool rotational speed, feed rate, and tool diameter. A quality weld joint can be produced if the K-factor is in the range of 150–400. Different kind of stresses is generated at the crown and root of the FSW seams. Bending type of stress arises in the welded region during the mechanical testing of FSW joints. The analysis shows that the crown side is exposed to compression whereas the root side of the weld seam undergoes tension (Kiss and Cziga, 2012). Here, different mechanical properties of the weldment such as tensile strength, shear bond strength, hardness, and impact strengths are discussed. The mechanical properties may vary based on the processing parameters.

Tensile Strength

The weld regions and the base materials behave differently to the loading conditions due to the varying microstructural features in the welded region and base materials. The reinforcement particles added in the polymer composite materials also affect the mechanical behavior of the FSW joints. Various sections of a tensile stress-strain plot for the HDPE composite base material is shown in Fig. 25(a). The stress-strain curve is similar to the characteristic behavior of semi-crystalline polymers. The uniform deformation is observed up to point A, and necking is initiated beyond point A. This is due to cold drawing and it propagates at constant stress level up to point B. This can be associated with stretching and realignment of the molecular chains in the direction of the applied load (Sheikh-Ahmad *et al.*, 2019). Tensile behavior is also affected by the discontinuity and non-uniformity of some defects such as tunnels and voids along the weld line. Fig. 25(b) shows the tensile behavior for specimens welded at 1000 rpm tool rotational speed and 30 mm/min tool traverse speed. The yield strength is reduced by 37% and the corresponding strain by 74% as compared to the base material (Sheikh-Ahmad *et al.*, 2019).

The fracture analysis will be useful in identifying the failure mode in the welded material. Fracture surfaces are examined using Scanning Electron Microscopy (SEM) after the tensile testing. The fracture analysis carried out with two materials having different yield strengths can provide a better understanding of the effect of material strength on the failure mechanisms. Two different

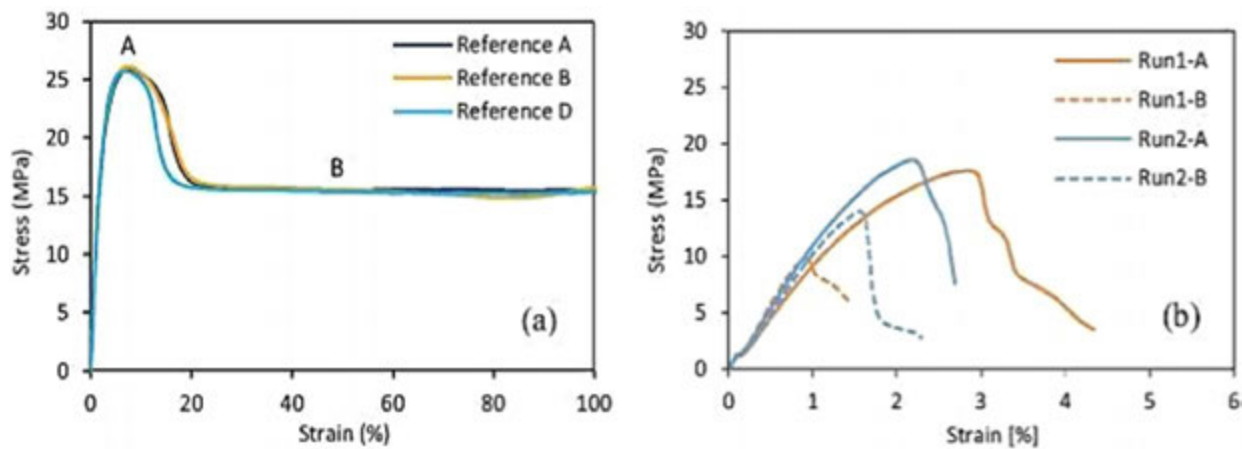


Fig. 25 Typical tensile test performance of the (a) base material for three reference samples (b) welded material. Reproduced from Sheikh-Ahmad, J.Y., *et al.*, 2019. Friction stir welding of high density polyethylene – Carbon black composite. *Journal of Materials Processing Technology* 264 (4), 402–413.

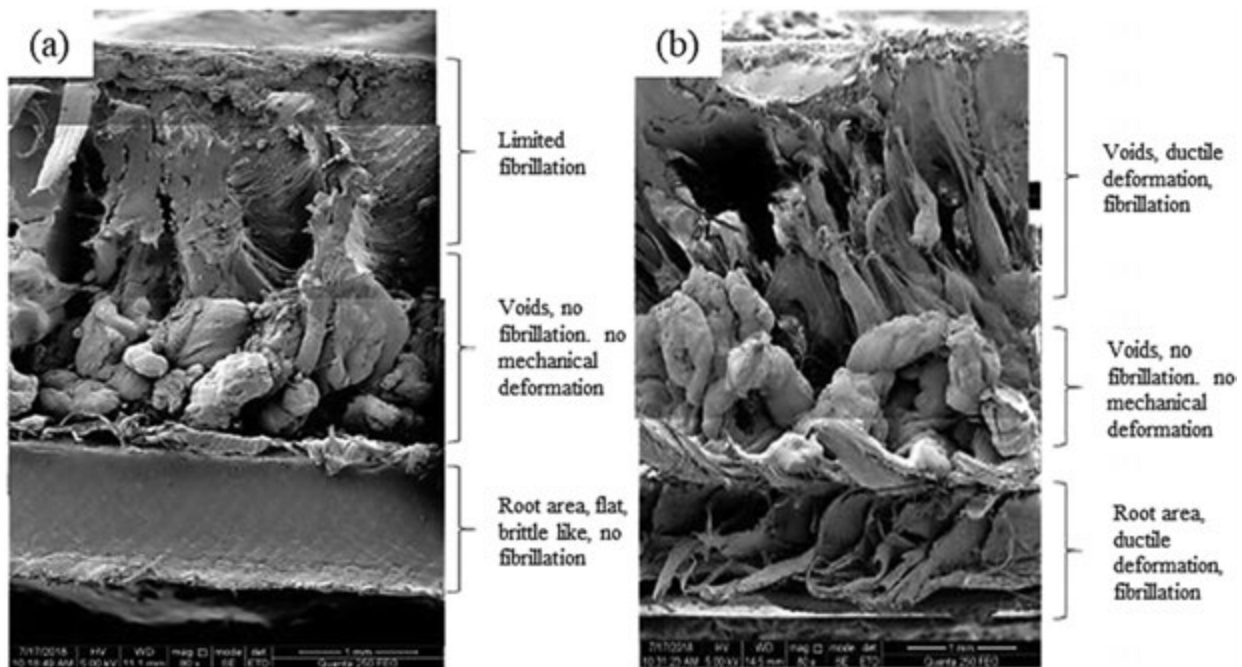


Fig. 26 (a) Macrostructure morphology of tensile fracture surface of sample 1 welded at 800 rpm, 20 mm/min (b) microstructure morphology of tensile fracture surface of sample 2 welded at 1000 rpm, 40 mm/min. Reproduced from Sheikh-Ahmad, J.Y., *et al.*, 2019. Friction stir welding of high density polyethylene – Carbon black composite. *Journal of Materials Processing Technology* 264 (4), 402–413.

specimens, one of low yield strength (4 MPa) and the other one of high yield strength (16 MPa) are analyzed as shown in Fig. 26 to understand the failure features of welded materials. The fracture surface of the low yield strength specimen shows a flat brittle area in the root region below the pin-influence zone. This is due to the lack of fusion between the two abutting workpieces at the root of the weld. The middle part of the fracture surface shows the morphology of a mixed material without fibrillation and the lack of mechanical deformation. This can be due to insufficient mechanical mixing at relatively low temperatures. Apparently, in this area, the material is not at all welded and it does not contribute to the tensile strength of the specimen. The upper part of the image presents limited ductility with minor fibrillations which contributes to the tensile strength of the sample. The material in that area is welded due to melting and solidification (Sheikh-Ahmad *et al.*, 2019).

The fracture surface for the high yield strength specimen is different than the low yield strength specimen. The ductile deformation at the bottom (root area) is identified in the fracture surface of the high yield strength specimen. This indicates that good root weld is obtained by forging at high temperatures which are obtained at the higher penetration depth.

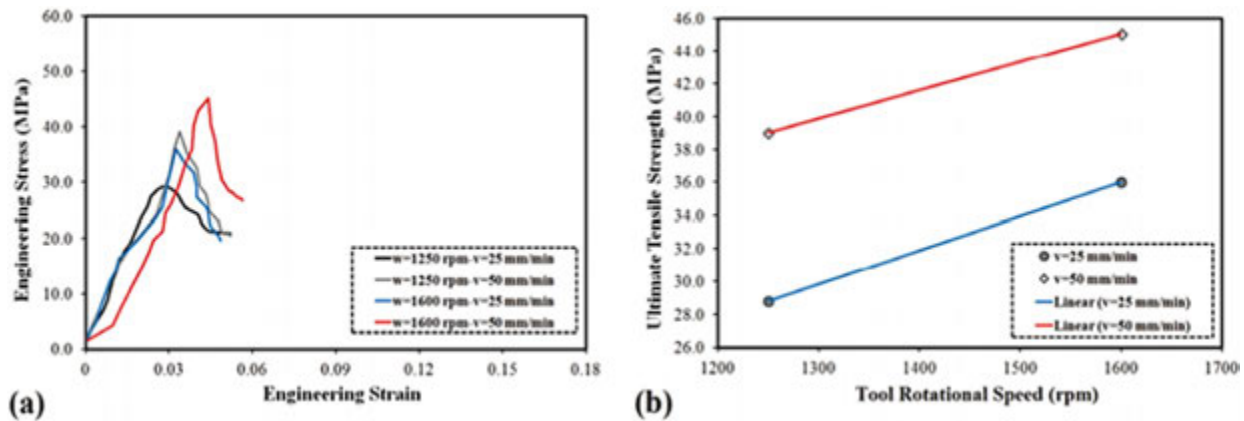


Fig. 27 (a) Engineering stress-strain curves for transverse tensile tested dissimilar joints. (b) Effects of rotational speed at different traverse velocity on the joint strength of dissimilar joints. Reproduced from Derazkola, H.A., Khodabakhshi, F., Simchi, A., 2018. Friction-stir lap-joining of aluminium-magnesium/poly-methyl-methacrylate hybrid structures: Thermo-mechanical modelling and experimental feasibility study. *Science and Technology of Welding and Joining* 23, 35–49.

A higher extent of material deformation is observed with cavities, most probably inherent from the welding process and enlarged during elongation. Unlike the low yield strength specimen, the mechanical deformation of this bottom area must be contributing to the tensile strength of the specimen. A mixed area without any fibrillation is observed in both specimens although the volume of the insufficient mixing area is less for the high yield strength specimen. In the middle region of the weld, the material does not show mechanical deformation. Therefore, no contribution to the tensile strength of the specimen is expected from this area. The upper part of the fracture surface shows much more elongated fibrils for the high yield strength specimen, which indicates higher ductility. Similar to the low yield strength specimen, the material in this region is welded by fusion due to melting. However, the extent of fusion is more in the high yield strength specimen. More tensile strength is also exhibited in combination with contribution from the root area (Sheikh-Ahmad *et al.*, 2019).

The maximum amount of heat generation during FSW is obtained for the high tool rotational speed and low tool traverse speed. The tensile strength of materials is affected by the generated heat input. The higher tensile strength is attributed to the better intermixing and interlocking at the higher heat input conditions (Derazkola *et al.*, 2018). The effect of processing parameters on tensile properties of dissimilar AA 5058 and poly-methyl methacrylate joint is shown in Fig. 27. At lower tool rotational speed, the decreased strength is due to the formation of a wormhole on the RS. At higher tool rotational speed, tunnel defect is observed due to excessive turbulence in the material flow. When the intermixing is insufficient at the RS, the material strength is reduced and the failure happens at the RS for the welded polymer matrix composites (Mendes *et al.*, 2014; Kumar *et al.*, 2019a).

Material flow and reinforcement content are two other factors that affect the tensile behavior of friction stir welded PMC. When two polymer workpieces are welded, the material flow leads to the intermixing of polymer phases. The heat generation at the RS is insufficient for the appropriate material flow. The insufficient material flow leads to limited intermixing and interlocking and thereby results in poor material adhesion. This leads to the initiation of crack at the RS (Laieghi *et al.*, 2019a). This kind of behavior is observed during the tensile test is performed for the Polyamide 6 and Nitrile butadiene rubber weld in the transverse direction. The addition of reinforcement particles help in increasing the tensile strength up to a certain extent. The increase in the reinforcement content above a certain limit leads to a decrease in the tensile properties of polymer composite-metallic material welding. The tensile strength values for lap welded short carbon fiber-reinforced PEEK and AA 2060-T8 at different tool rotational speeds are shown in Fig. 28 (Huang *et al.*, 2018b). The plot suggests that the tensile strength first increases and then decreases as the tool rotational speed increases as shown in Fig. 28(b).

The mechanical behavior of the friction stir spot welded polymers is different. The tensile force increases with applied strain for a certain value of strain and reduces upon further strain application. The force during the friction stir spot welding of polycarbonates increases linearly up to the maximum shear stress (F_s) as strain is applied on material as shown in Fig. 29. The corresponding displacement value is d_r . The amount of force required for the unit displacement is reduced as it crosses the d_r limit. A dramatic drop in the force takes place as the initial fracture completely loses the load-bearing capacity (Lambiase *et al.*, 2015).

The mechanical properties are also analyzed for the friction stir riveting of polymer composite materials. The failure of the rivet is tested and analyzed in two different ways: (1) pulling out the full rivet and (2) through the rivet. Insufficient mechanical anchoring in the substrate leads to poor intermixing and interlocking at the rivet surface. In such cases, the rivet tip is deformed and crack is initiated around the deformed rivet. This crack leads to pulling out the full rivet and leaves behind a hole similar to the diameter of the deformed rivet. The rivet also shows some elastic deformation and further the load is reduced due to the anchored rivet pulled out from the substrate. In the case of through the rivet failure mode, the fracture occurs outside the anchor zone. The extensive plastic deformation in PEEK reinforced with 30 wt% short carbon fibers is identified for both the failure modes as shown in Fig. 30 (Altmeyer *et al.*, 2015).

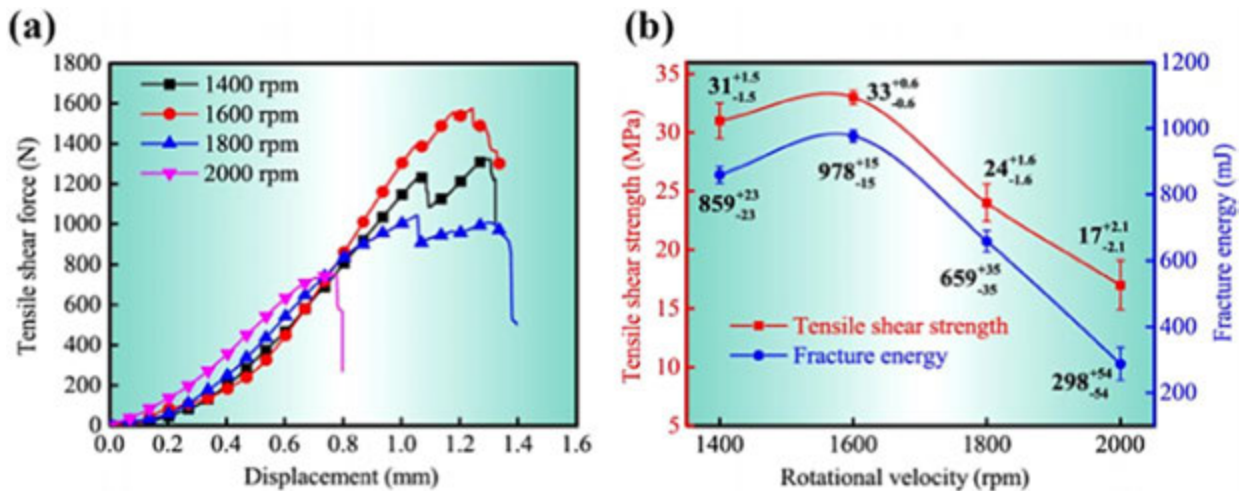


Fig. 28 (a) Force-displacement curves and (b) tensile shear properties of the joints. Reproduced from Huang, Y., Meng, X., Xie, Y., Li, J., *et al.*, 2018b. Joining of carbon fiber reinforced thermoplastic and metal via friction stir welding with co-controlling shape and performance. *Composites Part A: Applied Science and Manufacturing* 112, 328–336.

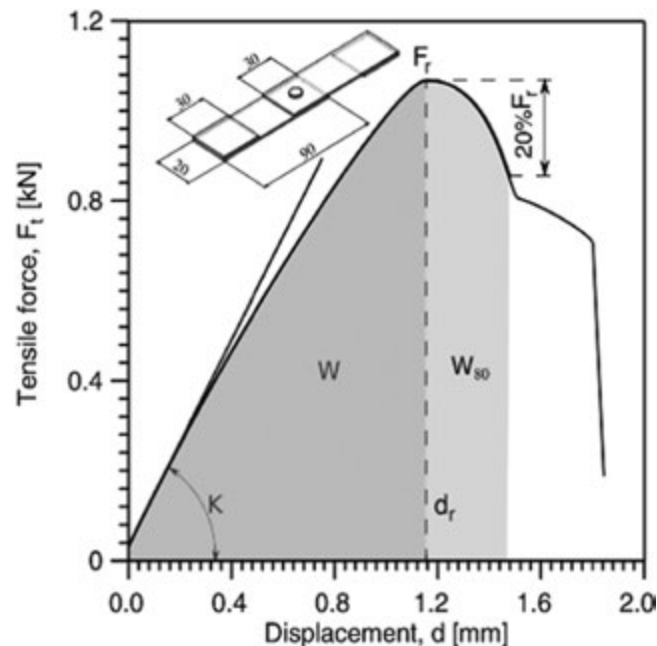


Fig. 29 Mechanical behavior of friction stir spot welded polycarbonates. Reproduced from Lambiase, F., Paoletti, A., Di Ilio, A., 2015. Mechanical behaviour of friction stir spot welds of polycarbonate sheets. *International Journal of Advanced Manufacturing Technology* 80 (1–4), 301–314.

The tensile strength of the materials is dependent on the process parameters, material flow, and reinforcement content in the polymer composite materials. Optimization of process parameters and reinforcement content can lead to the fabrication of welds with superior properties using FSW.

Shear Bond Strength

The shear bond strength of friction stir welds depend on the material flow during the welding. The material flow of metallic and polymeric materials is different which leads to the mixing of polymeric materials in the metal matrix in the form of anchors which enables the interlocking in the weld region. The higher shear strength is exhibited by bigger Al anchors in 6061-T6 aluminum (Al)- PEEK friction stir lap weld. The hardness is also improved with the higher Al anchor size as shown in [Fig. 31](#).

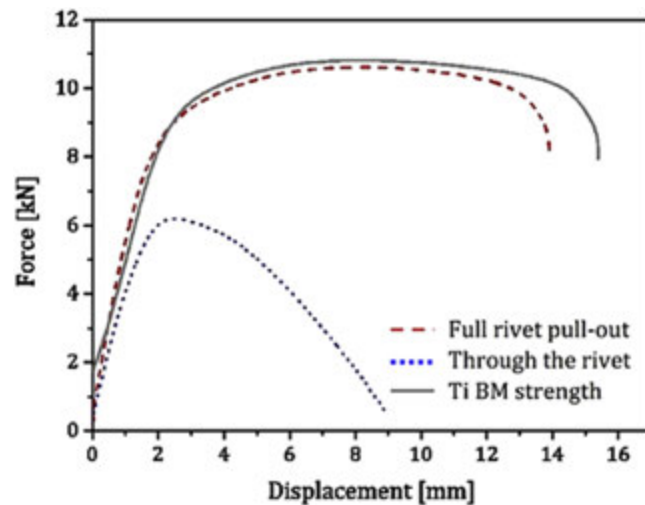


Fig. 30 Force-displacement curves comparing failure modes with the tensile strength of the grade 3 titanium base material rivet. Reproduced from Altmeyer, J., *et al.*, 2015. Microstructure and mechanical performance of metal-composite hybrid joints produced by FricRiveting. Composites Part B: Engineering 81, 130–140.

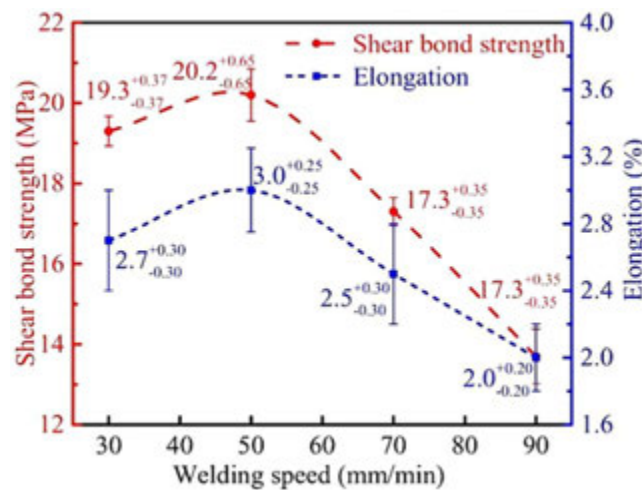


Fig. 31 Shear bond strength and elongation of the polymer-metal joints by different welding speeds. Reproduced from Huang, Y., Meng, X., Wang, Y., *et al.*, 2018a. Joining of aluminum alloy and polymer via friction stir lap welding. Journal of Materials Processing Technology 257 (9), 148–154.

The shear bond strength is higher when the gap at the interface between the molten and re-solidified polymer and metal is small (Huang *et al.*, 2018a).

Hardness

The hardness of friction stir welded polymer matrix composites is mainly influenced by the temperature generated based on the process parameter combinations as well as the type and amount of reinforcement particles. The maximum hardness is in the nugget zone which is followed by the advancing and retreating side, respectively. The addition of reinforcement materials such as Multi-Walled Carbon Nano-Tubes (MWCNT) above the critical amount reduces the hardness value due to a decrease in the destruction of the molecule structure (Gao *et al.*, 2015). During the FSW process, the molecular weight of the polymer is reduced due to high peak temperature and faster cooling which further leads to thermal degradation. At high tool rotational speed, the hardness is decreased due to higher heat generation. Therefore, the weld region which is prone to the higher heat generation exhibit lower hardness (Laieghi *et al.*, 2019a). The micro-hardness variation during the welding of AA 2060-T8 and PEEK with carbon reinforcement with respect to the tool rotation speed is shown in Fig. 32. The load-bearing capacity of the joint is dependent on the strength of the anchor that is formed during the lap welding. The hardness of aluminum anchor is also reduced

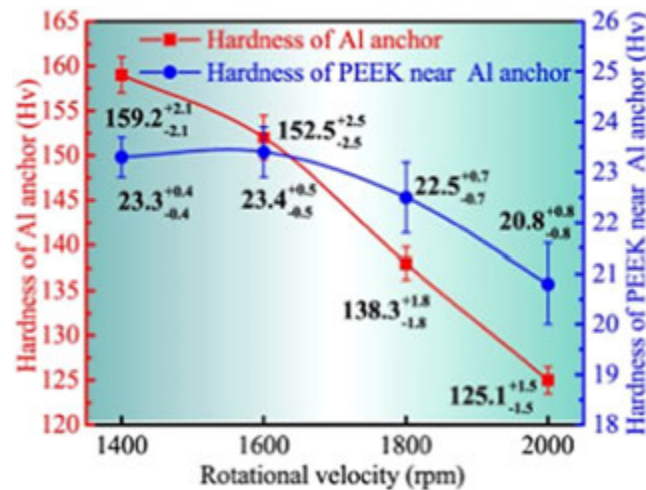


Fig. 32 Hardness of the typical locations for the Al anchor and its surrounding PEEK. Reproduced from Huang, Y., Meng, X., Xie, Y., Li, J., *et al.*, 2018b. Joining of carbon fiber reinforced thermoplastic and metal via friction stir welding with co-controlling shape and performance. *Composites Part A: Applied Science and Manufacturing* 112, 328–336.

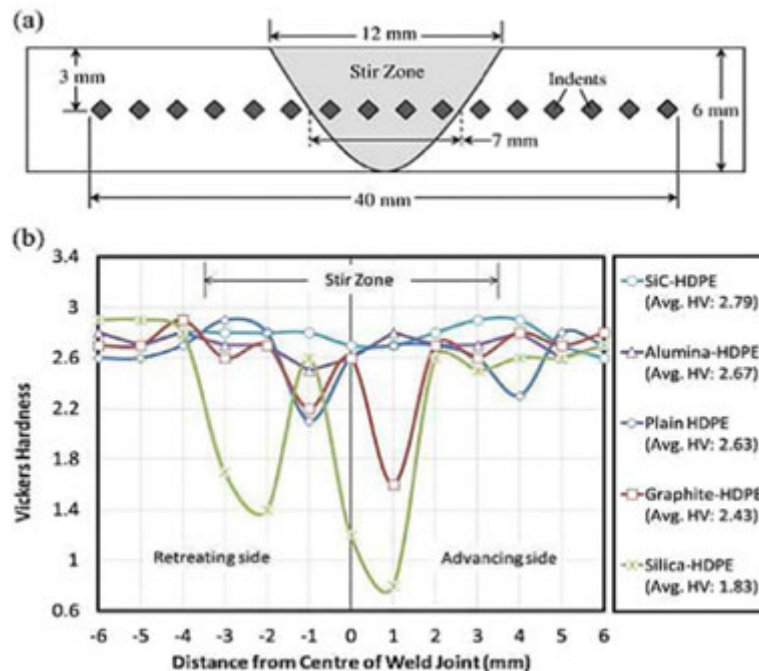


Fig. 33 (a) Schematic of the weld cross-section and indent location and (b) hardness profiles across weld nugget for different composite joints. Reproduced from Raza, K., *et al.*, 2017. On the friction stir welding, tool design optimization, and strain rate-dependent mechanical properties of HDPE–Ceramic composite joints. *Journal of Thermoplastic Composite Materials* 31 (3), 291–310.

with an increase in tool rotational speed. Deterioration of molecular weight and crystallinity lead to the drop in hardness surrounding the PEEK (Huang *et al.*, 2018b).

The hardness improvement in the FSW plain welded and composite joints of HDPE are analyzed and related to the efficiency of the FSW joints. Maximum joint efficiency (84%) is displayed by the plain welded specimens. The hardness plots for different types of additions and plain welds are shown in Fig. 33. The SiC reinforcement shows improved joint efficiency among different reinforcements (graphite, silica, and alumina). The particle size, morphology, and the amount of reinforcement particles also have influenced the end properties of the particle embedded HDPE matrix welds (Raza *et al.*, 2017).

Nanoindentation modulus and nanoindentation hardness of the welded HDPE-carbon black composite material is measured starting from the friction stir zone towards the base material along a line as shown in Fig. 34. The results have revealed that the nanoindentation modulus and hardness increase from the center of the weld towards the base material. Crystallinity is increased

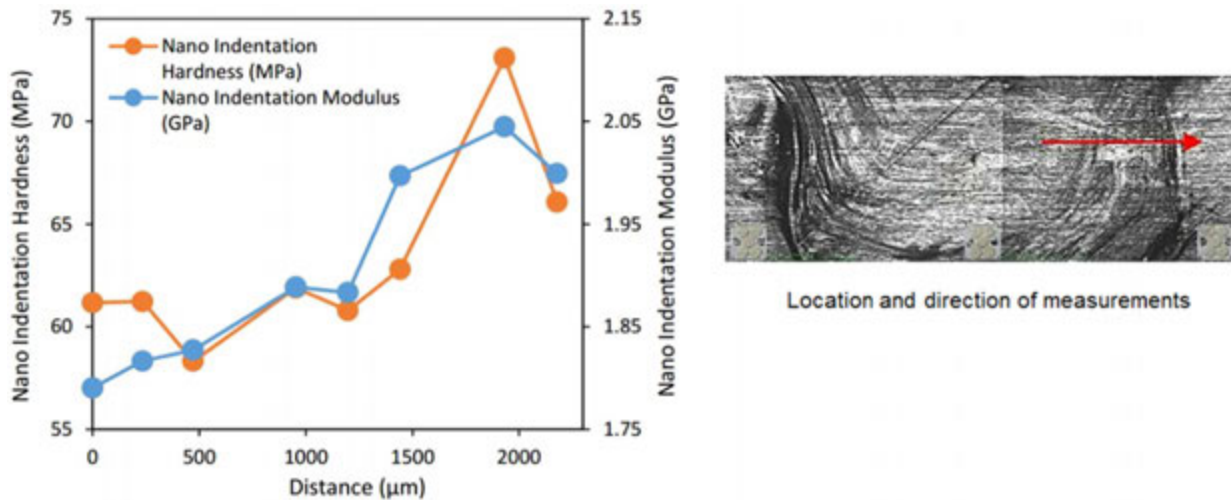


Fig. 34 Nanoindentation modulus and hardness of material along the red line. Reproduced from Sheikh-Ahmad, J.Y., *et al.*, 2019. Friction stir welding of high density polyethylene – Carbon black composite. *Journal of Materials Processing Technology* 264 (4), 402–413.

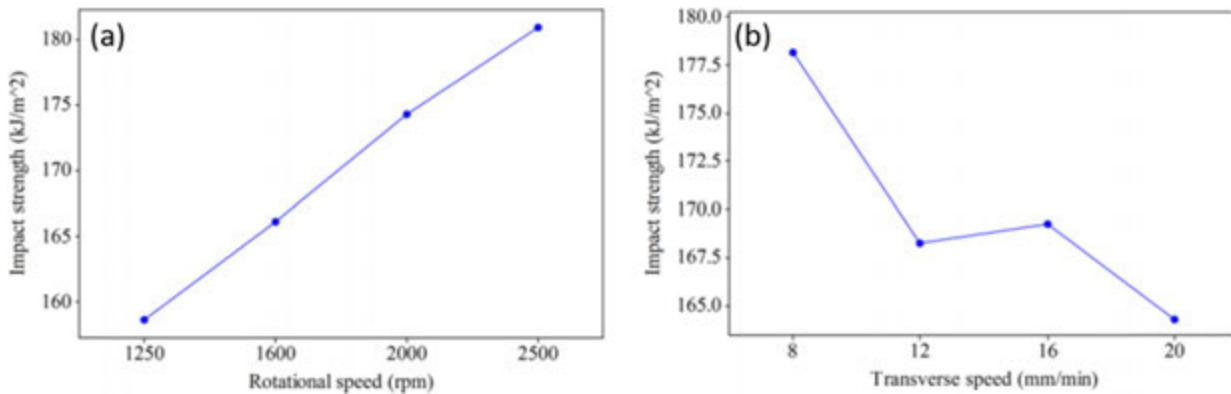


Fig. 35 Effect of (a) rotational speed and (b) traverse speed on impact strength. Reproduced from Azdast, T., Hasanzadeh, R., Moradian, M., 2018. Improving impact strength in FSW of polymeric nanocomposites using stepwise tool design. *Materials and Manufacturing Processes* 33 (3), 343–349.

from the center of the weld to the base material. The microvoids and cavities might have affected the nanoindentation test results to some extent. This could be the reason for the scatter of the readings (Sheikh-Ahmad *et al.*, 2019).

Impact Strength

The impact strength of friction stir welded polymers and polymer matrix composites depend on the processing parameters. Higher impact strength is observed for the higher tool rotational speeds. As the tool rotational speed is increased, the amount of heat generation also increases which further enhances the material softening. Therefore, the mixing of the base material is better at higher tool rotational speed. On the other hand, as the tool traverse speed is increased, the welding duration is reduced. As a result, material softening is less due to less interaction time. Therefore, the amount of material intermixing is reduced with an increase in tool traverse speed. Consequently, better impact strength is obtained at lower tool traverse speed (Azdast *et al.*, 2018). **Fig. 35** shows the impact strength of the polycarbonate weld with nano-alumina composites corresponding to tool rotational speed and traverse speed.

Defects

Appropriate welds can be formed in propitious weld materials for a certain range of welding parameters. Inappropriate heat generation during FSW results in the defects such as wormholes or voids, tunnels, surface grooves, excessive flash, collapsed nugget, surface galling, and kissing bond. The amount of heat generation is mainly governed by the tool rotational speed and

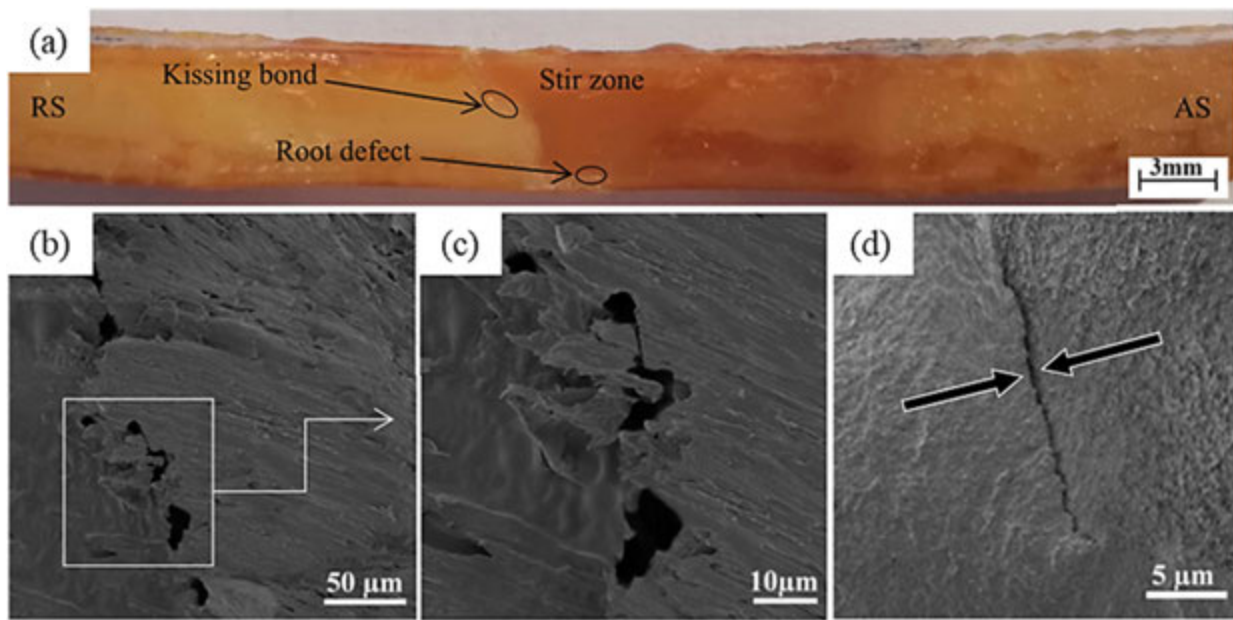


Fig. 36 SEM images showing the kissing bond and root defects in friction stir welded joint under high traverse speed. Reproduced from Laieghi, H., Alipour, S., Mostafapour, A., 2019a. Heat-assisted friction stir welding of polymeric nanocomposite. *Science and Technology of Welding and Joining* 25, 56–65. Available at: <https://doi.org/10.1080/13621718.2019.1610613>.

welding speed. The higher tool rotational speed results in excessive heat generation that leads to excessive flash formation. As a result, insufficient material remains to fill the weld surface which further leads to the surface groove defects. Lower tool rotational speed and higher welding speed results in insufficient heat generation. The defects such as voids and tunnels form due to limited material flow at lower heat input conditions. Abnormal stirring at the higher tool rotational and traverse speeds also lead to void defects. Sufficient heat generation and plastic deformation are obtained using a selected weld parameter range. Experimental limitations may also form defects such as lack of penetration.

The study on the general defects in friction stir welded polymeric materials is important to optimize the welding parameters for the successful weld. The parameters such as tool rotational speed and traverse speed affect the amount of heat generation. The amount of heat generation is higher at AS compared to RS as the shearing action takes place from the AS. Insufficient heat generation at the RS leads to void defects. In such cases, the kissing bond defects occur at the RS due to less adhesion between the stirring zone and base material. **Fig. 36** shows kissing bond defects identified for the higher traverse speed during the welding of polymer matrix nanocomposites. The lower thermal conductivity of polymer may cause incomplete root penetration, as shown in **Fig. 36(c)**. Such defects appear at the bottom of the joint when the material cannot be stirred with tool rotation. These defects can be minimized by optimizing the welding parameters (Laieghi *et al.*, 2019a).

The unfilled cavity defect occurs in the FSW of PEEK with short carbon fiber reinforcement and AA 2060-T8 at very low and very high tool rotational speed. At the lower tool rotational speed, the frictional heat is insufficient to fill the weld region. Therefore, cavity defect occurs as shown in **Fig. 37(a)**. The frictional heat generation and the plasticized material flow are enhanced at higher tool rotational speed. As a result, the plasticized material fills the cavity due to the material flow in the interface region as shown in **Fig. 37(b)**. When the tool rotational speed is too high, the maximum amount of polymer is spread in the joint region. Hence, an unfilled cavity is formed at the tool interface as shown in **Fig. 37(c)** and **(d)** (Huang *et al.*, 2018b).

The defect formation is also studied in the nugget zone during FSW of HDPE carbon-black composite. Different material regions are identified in the zone influenced by pin marked by letters A–C in **Fig. 38**. Flow lines are not observed in regions A and B of the base material. The material might have undergone complete fusion due to melting and resolidification with the base material. A large amount of voids on the top left quarter which is the pin-influenced zone shows the melting and material ejection. Voids on the bottom half of the pin-influenced zone are longer, aligned with the flow lines, and appear to be loaded in shear. These types of voids are observed to be dominant on the retreating side due to opposite flow directions of rotation and traverse (Sheikh-Ahmad *et al.*, 2019).

Numerical Analysis

Numerical modeling is used to study heat transfer and material flow during the friction stir welding of polymer matrix composites. The material flow in FSW is governed by complex physical phenomena which are difficult to study only using experimental techniques. Experimental analysis to understand these complex material behaviors will be cumbersome, time-consuming, and

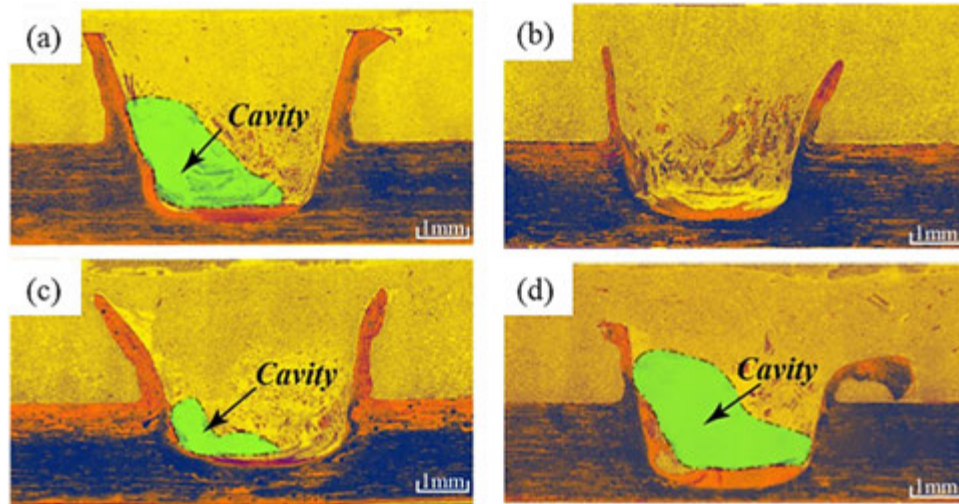


Fig. 37 Three-dimensional morphologies of the joints obtained by different rotational velocities (a) 1400 rpm, (b) 1600 rpm, (c) 1800 rpm, and (d) 2000 rpm. Reproduced from Huang, Y., Meng, X., Xie, Y., Li, J., *et al.*, 2018b. Joining of carbon fiber reinforced thermoplastic and metal via friction stir welding with co-controlling shape and performance. *Composites Part A: Applied Science and Manufacturing* 112, 328–336.

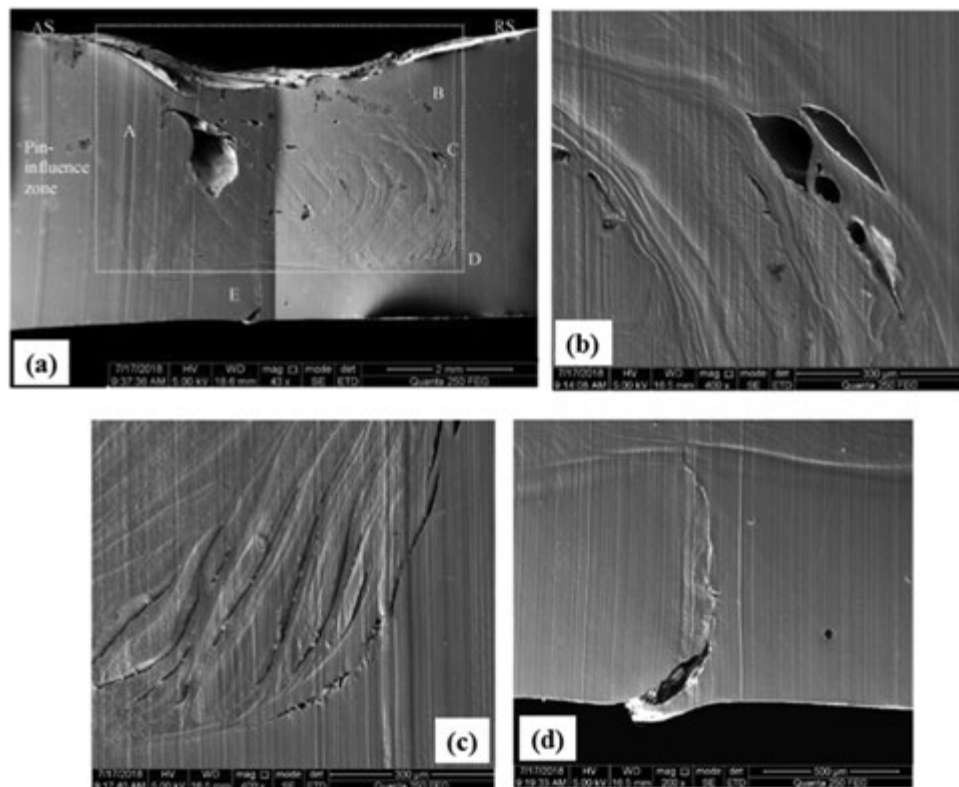


Fig. 38 SEM images of the cross-section of friction stir welded joint at conditions 1000 rpm rotation speed, 30 mm/min traverse speed and 4.1 mm plunge depth (a), details of voids at location C (b), details of voids at location D (c), root defect at location E (d). Reproduced from Sheikh-Ahmad, J.Y., *et al.*, 2019. Friction stir welding of high density polyethylene – Carbon black composite. *Journal of Materials Processing Technology* 264 (4), 402–413.

expensive. Numerical analysis is a solution to overcome such experimental shortcomings. Numerical analysis is preferred by many researchers to model heat transfer and complex material flow behavior. It can also test the feasibility of various process parameter combinations that are difficult to attain during an experimental plan.

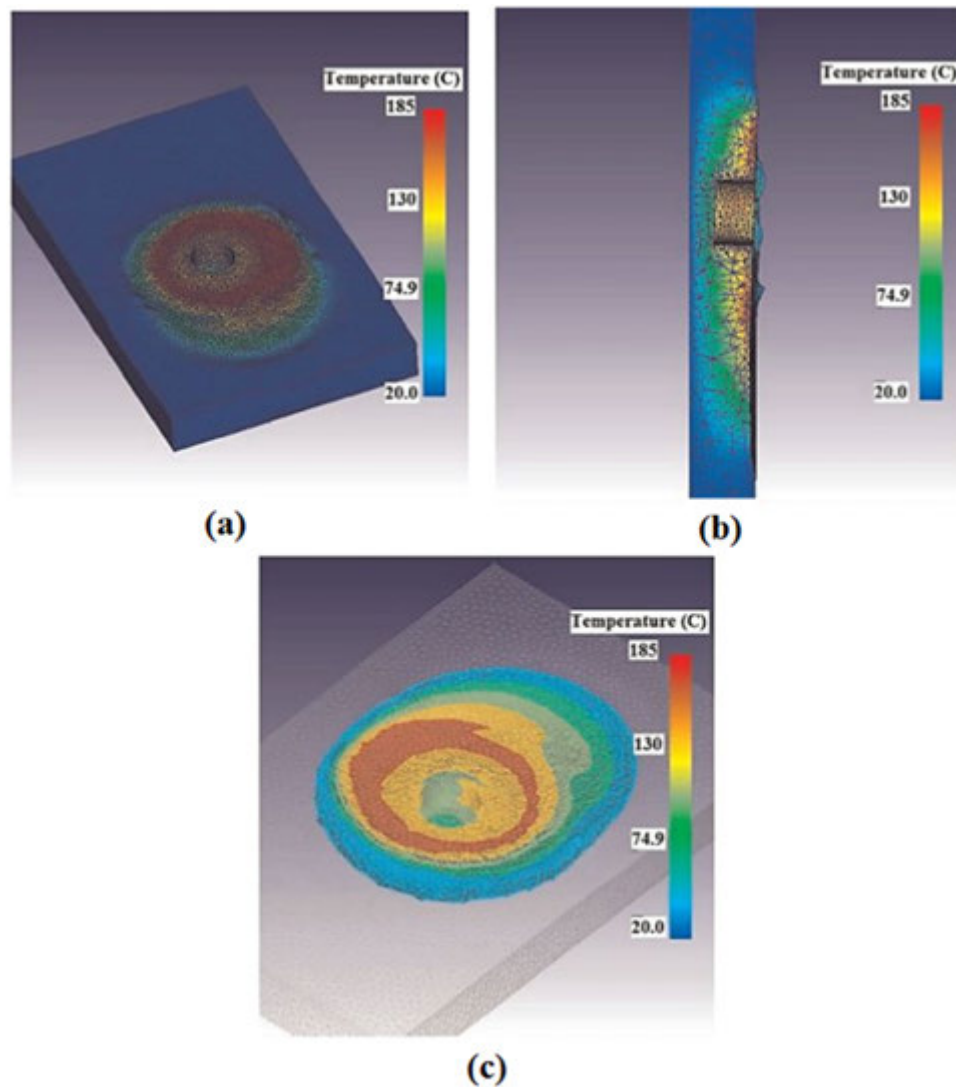


Fig. 39 Temperature contours along the tool path after 18 mm tool feed (a) at the surface, (b) in midline cross-section, and (c) in bulk. Reproduced from Zinati, R.F., Razfar, M.R., 2014. Finite element simulation and experimental investigation of friction stir processing of polyamide 6. Proceedings of the Institution of Mechanical Engineers, Part B: Journal of Engineering Manufacture 229 (12), 2205–2215.

The numerical model is used to predict the distribution of plastic strain and the material flow during the processing of Multi-Walled Carbon Nano-Tubes (MWCNTs) reinforced Polyamide (PA) 6 polymer matrix composite. The dispersion of MWCNTs in straight and con-curved fashion is confirmed by the numerical model and the actual experimental results. The thermal analysis concluded that the highest temperature is observed at the tool shoulder-workpiece interface and when the distance from the center of the tool is more, the temperature at the interface is less due to the asymmetric temperature distribution at the interface as shown in [Fig. 39](#) (Zinati and Razfar, 2014).

Numerical analysis is also used to model and understand the internal shear deformation mechanism and microstructure-dependent local buckling which is difficult to investigate through traditional 2D measurement techniques (Croom *et al.*, 2016). The new method combines micro-X-ray Computed Tomography (μ XCT) with Volumetric Digital Image Correlation (V-DIC) along with the in-situ mechanical testing to understand the 3D deformation behavior in the Friction Stir Blind Rivet (FSBR) joint of Carbon Fiber Reinforced Plastic (CFRP). This model has also illuminated the interfacial shear strain source between bulk material and the stir zone due to the compliant interface and heterogeneous stiffness in the stir zone (Croom *et al.*, 2016).

Numerical analysis can be carried out to understand the temperature distribution in the polymer materials using the inverse heat conduction method. [Fig. 40](#) shows the applied boundary conditions for inverse heat conduction method numerical analysis during the welding of HDPE carbon-black composite. Heat loss by convection and conduction is assumed at the top and bottom surfaces of the polymer composite, respectively. Different heat transfer coefficients are used to replicate both the altered conditions (Sheikh-Ahmad *et al.*, 2019). Temperature across the weld line increases sharply in a short distance, thereby possessing a higher temperature gradient. The heat conduction to both sides of the weld is very small due to the poor thermal conductivity of the

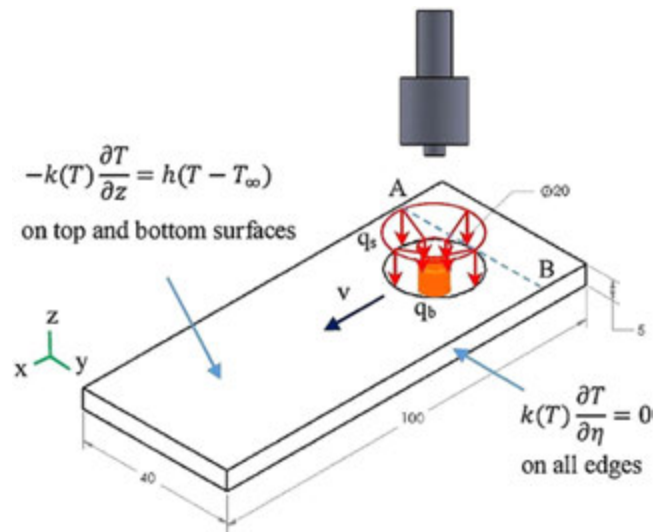


Fig. 40 Schematic for numerical modeling used in the welding of HDPE-carbon black composite workpieces. Reproduced from Sheikh-Ahmad, J.Y., *et al.*, 2019. Friction stir welding of high density polyethylene – Carbon black composite. *Journal of Materials Processing Technology* 264 (4), 402–413.

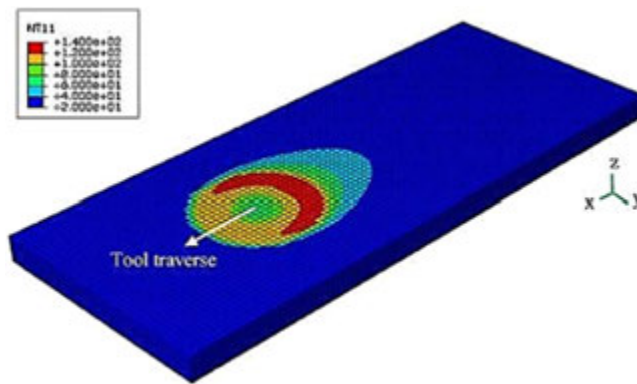


Fig. 41 Temperature distribution on the top surface of workpiece for welding speed of 30 mm/min. Reproduced from Sheikh-Ahmad, J.Y., *et al.*, 2019. Friction stir welding of high density polyethylene – Carbon black composite. *Journal of Materials Processing Technology* 264 (4), 402–413.

material. The temperature distribution is symmetrical on both sides of the weld line. Various zones during welding where melting of the material happen are visualized with the help of numerical analysis. The melting of materials majorly happens at the trailing side of the tool pin and underneath the tool shoulder.

Fig. 41 shows the temperature distribution on the top surface of the HDPE carbon-black composite workpiece. Thermal profiles on the bottom surface show that the temperatures under the tool shoulder are relatively higher than those in pin surrounding region and areas distant from the shoulder. Heat accumulation at the trailing shoulder surface shows that this region possesses the highest surface temperatures (Sheikh-Ahmad *et al.*, 2019).

Fig. 42(a) and **(b)** show the temperature distribution on a transverse cross-section passing through the center of the pin and longitudinal cross-section in the middle of the workpiece, respectively. Shoulder temperatures are higher than the other regions of the weld zone. At the trailing end of the shoulder, heat accumulation will be more up to a small depth. Since the material is not transferred to the pin-influenced zone by the cross-flow, it is ejected outside the weld zone by the centrifugal force of the shoulder. The central pin region shows a variation of temperature from the trailing side of the pin to the leading side and from top to bottom. The pin rotation makes the molten material flow from the trailing edge to the advancing side and then it is partially ejected from the stir zone. As shown in **Fig. 42**, this leads to the formation of large voids like wormholes and tunnels (Sheikh-Ahmad *et al.*, 2019).

The numerical analysis helps understand the complex mechanism of heat transfer and material flow during the welding and joining of polymer materials and polymer matrix composites.

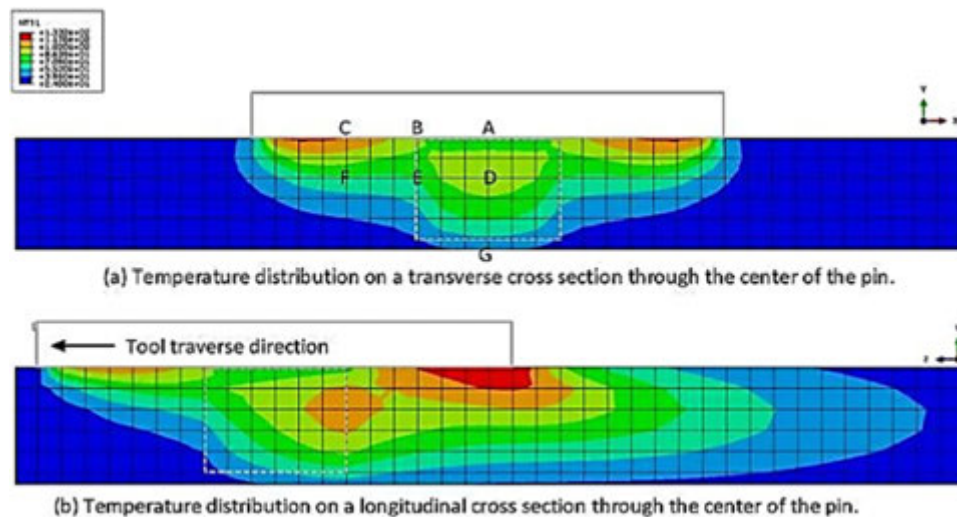


Fig. 42 Temperature distributions on longitudinal cross-section. Reproduced from Sheikh-Ahmad, J.Y., *et al.*, 2019. Friction stir welding of high density polyethylene – Carbon black composite. *Journal of Materials Processing Technology* 264 (4), 402–413.

Summary

High strength-to-weight ratio makes polymer materials and PMCs a suitable candidate for automotive applications. The conventional welding techniques are not suitable in the joining of polymers and PMC materials due to the lower melting temperature of the matrix material. FSW is a solid-state welding technology that can be used for the welding of polymer based materials. The peak temperature during FSW is close to the melting point to ensure the partial melting and mechanical mixing of the polymers for a successful joint formation. The properties of the polymeric chains are retained as the complete melting of the polymers is avoided during FSW.

FSW tool with stationary or rotating tool shoulder, with or without external heating, are used for the FSW of polymer based materials. Submerged welding, friction stir spot welding, and friction riveting are some innovative variations of the conventional FSW used for the welding of polymers and PMCs. FSW of PMCs is carried out in lap and butt configurations. Tool rotational speed, tool traverse speed, tool tilt angle, tool geometry, and reinforcement content influence material flow and microstructure during FSW of polymers and PMCs. These welding parameters also affect the mechanical properties such as weld strength and hardness, and determine occurrence of welding defects. The numerical analysis of material flow and heat transfer during welding helps to overcome the experimental limitations during the study on the welding of PMCs. The physical phenomena involved in PMC welding are complicated and a comprehensive understanding of the weld zone behavior is necessary. The fully homogenized reinforcement particle distribution during PMC welding is yet to be achieved. The detailed conceptual understanding of the effect of advanced tool geometries, optimized processing parameter combinations on the weld properties is important and an interesting field of research.

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<https://amit.people.iitgn.ac.in/>
 Amit Arora.
 IIT Gandhinagar.
<https://www.twi-global.com/>
 TWI Global.

Joining of PMC to Concrete for Structural Applications

Jesús Justo, Alberto Barroso, Antonio Blázquez, and Federico París, Sevilla University, Seville, Spain

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Introduction

The properties and features of concrete and composites are very well known. On the one hand concrete is a material with satisfactory behavior under compression and almost no strength under tension, with a very low price and remarkable durability and low requirements for maintenance. On the other hand, composites present excellent properties under tension and similar under compression provided that local and global lack of stabilities are avoided. In contrast, the price is extremely high and preserve a reasonable level of properties in terms of durability and maintenance (absence of corrosion).

The role of composites, due to their very low specific weight, has been in its origins, confined to applications of mobile structures in the fields of aircrafts, cars, bikes or boats, among many others, the weight playing in all these fields a determinant role.

Concrete has been traditionally used in applications where the weight plays almost no role at all. In spite of this, there is a clear complementarity between these two materials in applications where the features of both can be advantageously used.

Rehabilitation through structural strengthening is, with no doubt, the field where composite materials have found the aforementioned complementarity both for masonry and concrete structures. The extremely high properties of strength of composites suggest to be used in a small scale in comparison with concrete, the price not representing then a serious restriction in this case.

Rehabilitation is often used in structures that are damaged, poorly executed or simply, in absence of previous factors, because the structure may suffer a change on its use, then requiring a greater carrying capacity.

The operations required in rehabilitation entail in-situ reinforcement that very often involve in turns a high capacity of adaptation to a huge variety of structural shapes. This is precisely one of the excellent properties of composites, as is immediate to recognize that the 3D printing technique, of high predicament nowadays due to its capacity to generate all types of structural shapes, is just an extension of the technique of fabrication of composites by means of deposition of layers (Crosky *et al.*, 2015; Elkington *et al.*, 2015; Kim *et al.*, 2014; Olsen, 1993; Raspall *et al.*, 2019; Szczesny *et al.*, 2017).

It is immediate to notice that independently of the problems associated with each of the components involved in rehabilitation, a key problem will be the bonding between concrete and composite, although this is not a problem exclusive of the application under consideration in this chapter. In fact, this is one of the most serious problem that still remain to be solved in other applications of composite as for instance the aeronautic industry. When the substitution of the mechanical joining of composites by adhesive bonding was expected to happen, the use of adhesive bonding is still very restrictive, particularly in the case of primary structures of aircrafts. The lack of a procedure to elucidate the quality of a bonded joint has limited the applicability of this technique and constitutes an object of study of first level (see, for example, the Cost Action CA18120 - *Reliable roadmap for certification of bonded primary structures*, funded by the Horizon 2020 Framework Program of the European Union). Obviously, this problem arises here aggravated by the poor conditions of the surfaces involved in the bonding, particularly that of concrete, as well as the requirements in the control of the bonding procedure due to the in-situ limitations that typically appear. There are, in the field object of this chapter, few regulations in terms of the tests required, see for instance those of the Italian National Research Council (CNR-DT200, 2013) or the Fédération Internationale du béton (Triantafillou *et al.*, 2001).

There are many factors that may influence the properties and performance of a concrete-composite bonded joint. Thus, in connection with the concrete substrate, the treatment of the surface and the own strength of the concrete (Iovinella *et al.*, 2013; Toutanji and Ortiz, 2001) are clearly key factors. In connection with composites, factors like the type of resin and fibers, the stacking sequence, the manufacturing technique and the curing temperature (Czaderski *et al.*, 2012) must be mentioned. Finally, in connection with the adhesive, it is clear that the manufacturing technique of the joint (Jumaat *et al.*, 2011) as well as the properties of the own adhesive are influencing factors. There are in addition factors like the temperature and the moisture that must also be taken into account (Wang *et al.*, 2020; Silva *et al.*, 2013).

To characterize a bonded joint is not an easy task as there may be different mechanisms of damage involved in the final failure. Thus, Justo and París (2018), we can have damage in form of cracks in the concrete, damage in the composite in form of breaking of the fibers or delamination between laminas of the laminate. We can also have failure in the adhesive which is typically known as cohesive failure, or failure between the adhesive and concrete or composite which is known as adhesive failure. A well conceived and executed joint must imply the breakage of the concrete, as the weakest part of the whole joint.

The performance of tests, in order to get more than an experimental knowledge about the mechanisms of damage involved in the failure of the joint, must be accompanied by numerical simulations, in order to generate information about the properties implicated in the different mechanism of damage detected in the failure. This will help in having a tool for helping in the design of safe and efficient joints. A particular concern is referred to the fact that in the modeling of some of the tests that must be carried out, there are nominal singular stress state involved, which requires the use of very refined tools to describe such complex, from a mathematical point of view, situations.

In this chapter, Section "Materials and Methods" describes the materials used as well as the tests that are going to be carried out. The inspection of the geometry of the tests identifies, as already mentioned, the presence of corners with singular stress state which

are prone to unchain the failure. The problems associated with these situations are described in Section “Materials and Methods” as well as the numerical technique employed in the simulation of the tests carried out.

Section “Tests on Cylindrical Trepanned Coupons. Pull off and Shear Torsion Tests” treats jointly the pull off and shear torsion tests whereas Section “Tests on Cantilever Coupons” treats jointly lap shear and lap peeling tests. Both sections are organized in a similar way. First the tests are described with the corresponding identifications of associated problems. Then the preparation of the specimens and the experimental results found are shown. The virtual testing, each case with the associated problematic, is presented, finishing with a discussion on the experimental correlations between predictions and experimental results.

A general section of conclusions closes the chapter.

Materials and Methods

Materials Under Study

Three materials have been studied in this chapter, the two adherents, composite and concrete, and the adhesive, whose properties are described in what follows.

Composite material: formed by epoxy resin as the matrix and carbon fibers as the reinforcement. The resin is intended to be cured at room temperature (with a fast and improved cure at 60°C) and has 3.5 GPa of tensile modulus and 85 MPa of tensile strength. The fibers have 230 GPa of tensile modulus and 4900 MPa of tensile strength. All these values refer to nominal values of the corresponding property.

Concrete: the concrete blocks used in the test campaign are formed by H-35 concrete, which has a characteristic strength of 35 MPa.

Adhesive: the adhesive used is the same resin used in the composite, with 3.5 GPa of tensile modulus and 85 MPa of tensile strength.

Problems Under Study

To study the quality of the joint, four tests are considered in this work: pull off, shear torsion, lap shear and lap peeling. The tests are described next in what follows.

- (1) **Pull off:** the goal of this test is to submit the specimens to a tensile load perpendicular to the joint interface, trying to pull off the composite laminate from the concrete. The specimen consists on a circular coupon that has been trepanned. In order to apply the load, a metallic block is bonded to the composite layer. A scheme of the test is shown in [Fig. 1](#).
- (2) **Shear torsion:** the objective of this test is to bring the joint under a shear state, applying a torsion load. The specimen, again, consists on a circular coupon, with a bonded metallic block, that has been trepanned. A schematic view of the test can be seen in [Fig. 2](#).
- (3) **Lap shear:** in this test, a shear state is created in the joint by the application of a tensile load to the composite laminate in the plane of the laminate. In this case, the coupon consists on a concrete block where a composite laminate is bonded. Part of the laminate remains out of the block, in order to allow the load to be applied. In this case, the concrete block is fixed and cannot scroll or rotate. An outline of the test is shown in [Fig. 3](#).
- (4) **Lap peeling:** peeling stresses at the joint are generated in this test. To this end, a load is applied to the composite laminate in the direction of the thickness. The specimen is the same than in the case of the lap shear test. The concrete block can be moved freely in the longitudinal direction of [Fig. 4](#), where an outline of the test is shown.

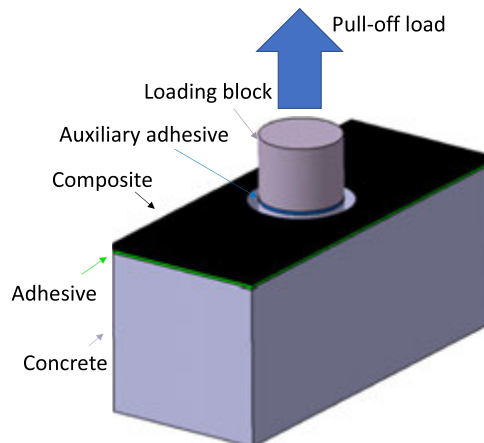


Fig. 1 Scheme of the Pull off test.

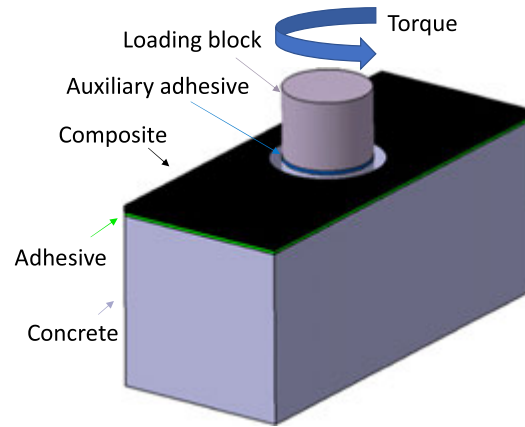


Fig. 2 Scheme of the shear torsion test.

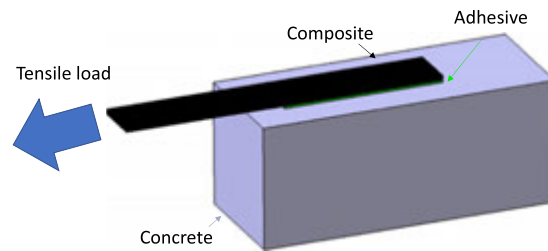


Fig. 3 Scheme of the lap shear test.

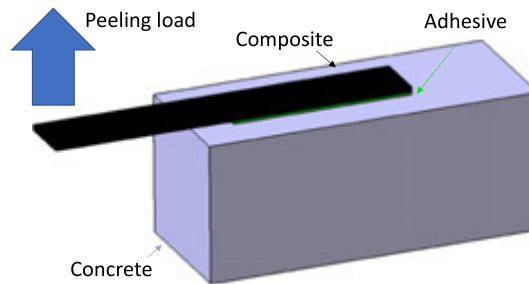


Fig. 4 Scheme of the lap peeling test.

It can be seen that Pull off and shear torsion tests use the same kind of coupon and share part of the problematic associated to it. The same fact happens for lap shear and lap peeling tests. Because of this and for the sake of clarity, the tests will be grouped in Sections "Tests on Cylindrical Trepanned Coupons. Pull off and Shear Torsion Tests" and "Tests on Cantilever Coupons", respectively, presenting together the characteristics they have in common.

Stress Singularities at Multimaterial Corners

The experimental campaign to be done implies testing until failure of the specimens subjected to different loading conditions. The presence of different materials in the samples generate critical points, where the failure is prone to occur. The abrupt change in the geometry and in the material properties, creates unbounded stress states at these critical points (from a linear elastic point of view).

The previous experience ([Barroso et al., 2020](#)) with the premature failure of samples where different materials are bonded using adhesive, with the failure starting at these corners, has encouraged the authors to make a singularity stress analysis of the different multimaterial corners appearing at the test samples to check if these singularity stress fields have any influence in the failure of the samples.

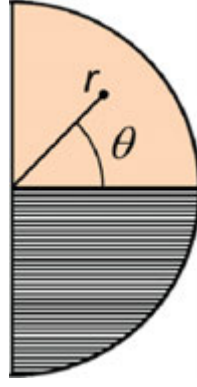


Fig. 5 Polar coordinate system at the material corner.

Taking a polar coordinate system (r, θ) with the origin at the corner tip (see for example [Fig. 5](#) with a bimaterial corner), the asymptotic singularity stress field can be represented by the expression:

$$\sigma_{\alpha\beta}(r, \theta) = \sum_k K_k r^{-\delta_k} f_{\alpha\beta}^{(k)}(\theta) \quad (\alpha, \beta = r, \theta) \quad (1)$$

in which, K_k are the Generalized Stress Intensity Factors (GSIFs), $0 < \delta_k < 1$ are the order of stress singularities, and $f_{\alpha\beta}^{(k)}(\theta)$ are the characteristic angular shape functions ([Barroso et al., 2003](#), [Mantič et al., 2013](#)), δ_k and $f_{\alpha\beta}^{(k)}(\theta)$ depend on the local geometry, local material properties and local boundary conditions (close to the corner tip), while K_k depend on the global geometry and far field loading. There are few particular cases where the stress representation in [Eq. \(1\)](#) present logarithmic terms, for further information see ([Sinclair, 1999](#)). In the present work we will only characterize the orders of stress singularities (δ_k) as an estimation of the stress severity at each multimaterial corner.

The stress characterization will be carried out using a software developed by the authors ([Barroso et al., 2003](#), [Mantič et al., 2013](#)). With this tool, implemented in Mathematica, multimaterial corners of any number of materials can be analyzed. The materials can be isotropic, orthotropic or general anisotropic, and are assumed to be perfectly bonded between them (although the code also allows frictional interface between the materials). The boundary conditions at the external faces can be free, fixed, symmetry, or any combination of the polar variables of stresses and/or displacements.

The local geometry, local boundary conditions and local material properties will be detailed in the corresponding section, for each test configuration, together with the details of the stress singularity results, and failure analysis.

Numerical Techniques

The numerical simulation of the tests implies the modelization of the damage progression across the interface. Nowadays, there are several tools available: virtual crack closure technique (VCCT: [Agrawal and Karlsson, 2006](#), [Xie and Biggers, 2006](#)), cohesive zone models (CZM: [Turón et al., 2007](#); [Reinoso et al., 2017b](#)), continuum damage models (CDM: [Ladevèze and Le Dantec, 1992](#); [Reinoso et al., 2017a](#)), linear elastic brittle interface model (LEBIM, [Mantič et al., 2015](#); [Távora et al., 2019](#)), extended FEM (X-FEM: [Moës et al., 1999](#); [Linder and Armero, 2007](#)), and Phase Field (PF: [Pham et al., 2011](#); [Quintanas-Corominas et al., 2019](#)).

In the numerical analyses performed in this chapter the CZM implemented in Abaqus software has been used. Prior to the initiation of the damage, the behavior of the bonded interface is described as linear elastic stiffness until the traction reaches a maximum, then degrading progressively (several degradation laws are proposed by the scientific community) until the components become completely detached. [Fig. 6](#) shows a classical traction-separation law with linear degradation used for fracture mode I (a) and fracture mode II (b). The shadow areas represent the G_{IC} and G_{IIC} properties involved in the problem. The relative proportions of normal and shear deformation in the joint is quantified by the mode mixity. A particularly useful fracture criterion to account the mixity is the Benzeggagh-Kenane fracture criterion ([Benzeggagh and Kenane, 1996](#)) that defines a specific G_C depending on the ratio of $G_{II}/(G_I + G_{II})$ and a parameter η that should be adjusted experimentally:

$$G_C = G_{IC} + (G_{IIC} - G_{IC}) \left(\frac{G_{II}}{G_I + G_{II}} \right)^\eta \quad (2)$$

Tests on Cylindrical Trepanned Coupons. Pull off and Shear Torsion Tests

Test Description

For the pull off and shear torsion tests, a CFRP laminate is bonded to the concrete block. Then several cylindrical trepanations are performed and a metallic block is additionally bonded at each trepanation cylinder to apply the load. For the pull off test a

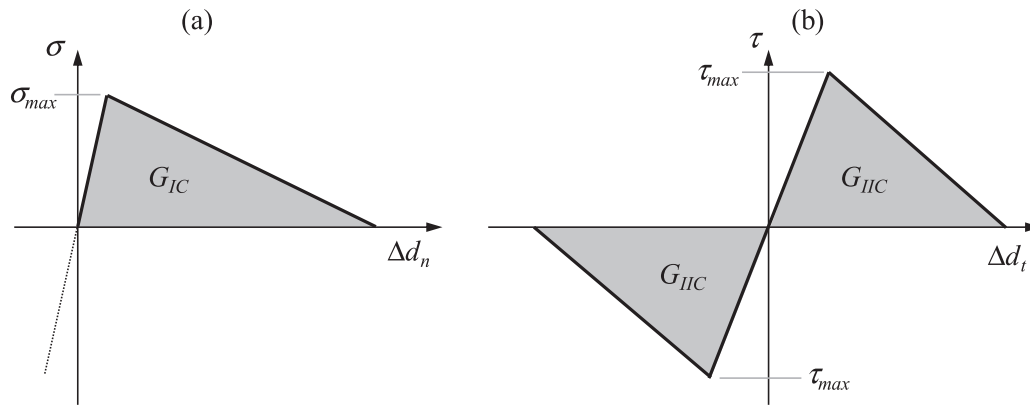


Fig. 6 Cohesive zone model for (a) mode I and (b) mode II.



Fig. 7 Pull off test device and samples.

traction load will be applied perpendicular to the surface, while in the shear torsion test a torque will be applied, as was previously introduced (Section “Problems Under Study”). The adhesive used to bond the metallic loading block to the composite laminate is different from that used to bond the composite laminate to the concrete. The first adhesive has been chosen with better mechanical properties than the second one, in order to avoid undesirable failures outside the area of interest.

The pull off load is applied by means of a Proeti pull off strength device, depicted in [Fig. 7](#), which is typically used in this type of tests to measure the strength of the adhesive joint to the concrete block. The diameter of the circumferential trepanation is 54 mm in the internal side and 60 mm in the external side (the thickness of the trepanation crown is 3 mm). The depth of the trepanation is 15 mm and the thickness of the composite laminate is 2 mm, thus the depth of the trepanation in the concrete is 13 mm. The height of the auxiliary metallic block to apply the pull off load is 35 mm.

The test is performed according to ASTM D4541 ([ASTM D4541-17, 2017](#)). The load is applied manually and the maximum load is recorded by the device. Due to the characteristics of the testing device, a load-displacement curve is not available (just the maximum load value).

For the shear torsion test, the trepanation procedure and the metallic block bonding are exactly the same. The load is applied by means of a torque wrench, as depicted in [Fig. 8](#). Notice that the test fixture also introduces a force in the sample, but, due to the ratio of the moment arm distance to the sample diameter, the effect of this force can be considered negligible compared to the torsion introduced in the sample.



Fig. 8 Shear torsion test fixture and samples.

Table 1 Experimental results for the pull off tests

Surface treatment	Manufacturing procedure								
	Pre-cured			Hand lay-up			Infusion		
	Load (N)	Std. Dev (N)	Cov (%)	Load (N)	Std. Dev (N)	Cov (%)	Load (N)	Std. Dev (N)	Cov (%)
Grinder	4969 (20%)	1830	36.83 ^{2,3}	4125 (ref)	1806	43.79 ³	7148 (73%)	1212	16.96 ²
Grit sandpaper	5121 (24%)	853	16.67 ¹	7621 (84%)	933	12.25 ¹	6630 (60%)	1190	17.96 ¹

The torque wrench does not continuously measure the applied torque. A maximum torque value to transmit by the torque wrench can be set and this procedure is done in discrete steps of 5 Nm. The maximum torque is obtained as the mean value between the last two consecutive values set at the torque wrench before failure.

Coupons' Preparation and Experimental Results

Three different manufacturing procedures and two different surface preparations were applied to bond the laminate on the concrete block. For the manufacturing process, the procedures used were: (1) bonding of a precured laminate, using the same resin of the composite material at 60°C for 24 h under a constant pressure of 0.28 MPa, (2) in situ lamination, with a hand lay-up of uncured laminas of the composite material on the concrete block, using the same curing parameters as detailed before, and (3) resin infusion, using dry fiber placed over the concrete block and a vacuum bag to carry out the infusion and bonding with the resin, using the same curing parameters than in the other two previous methods. For the surface treatment, grinder and grit sandpaper were used as alternative procedures.

Trepanation of the samples was performed using a tungsten carbide circular crown with intensive water cooling to avoid the burning of the resin. 5 samples were tested for each combination of manufacturing procedure (3) and surface treatment (2), thus involving a total of $3 \times 2 \times 5 = 30$ samples.

Test results for the pull off test are summarized in [Table 1](#), in which the value of the mean maximum load is presented for each configuration. The lowest observed value has been taken as reference (4125N), which corresponds to the combination of hand lay-up + grinder. The values between parentheses in [Table 1](#) show the % of failure load increment obtained in the tests in comparison with the reference value.

The superscripts in the value of the Covariance indicate the location where the sample was trepanned in the concrete block. [Fig. 9](#) illustrates: (1) the different locations where the samples were obtained from, (2) a detail of the failure, and (3) some broken samples of the pull off test.

It is noticeable that all samples failed inside the concrete zone. Some of them near the bottom of the trepanation depth (15 mm), but the majority of the samples at different depths.

The samples prepared with the grinder showed a high dispersion in the failure load values, whereas lower dispersion was found in the samples prepared with the grit sandpaper.

The results in [Table 1](#) show a high influence of the manufacturing procedure in the failure load, which could be surprising, as all samples broke inside the concrete (thus, with no possible influence of the way the laminate was bonded to the concrete). The explanation for that effect is associated with the location where the samples were prepared. In [Fig. 9\(a\)](#) some samples were prepared at the lowest face of the block (zone 1) and others at the lateral face (zones 2 and 3). The highest load and lowest dispersion was found in samples belonging to zone 1, then lowest failure load values were observed in samples belonging to zone 3. The lower the location of the samples in the concrete block, the higher the failure load observed, due to a better compaction and more homogeneous preparation of this zone.

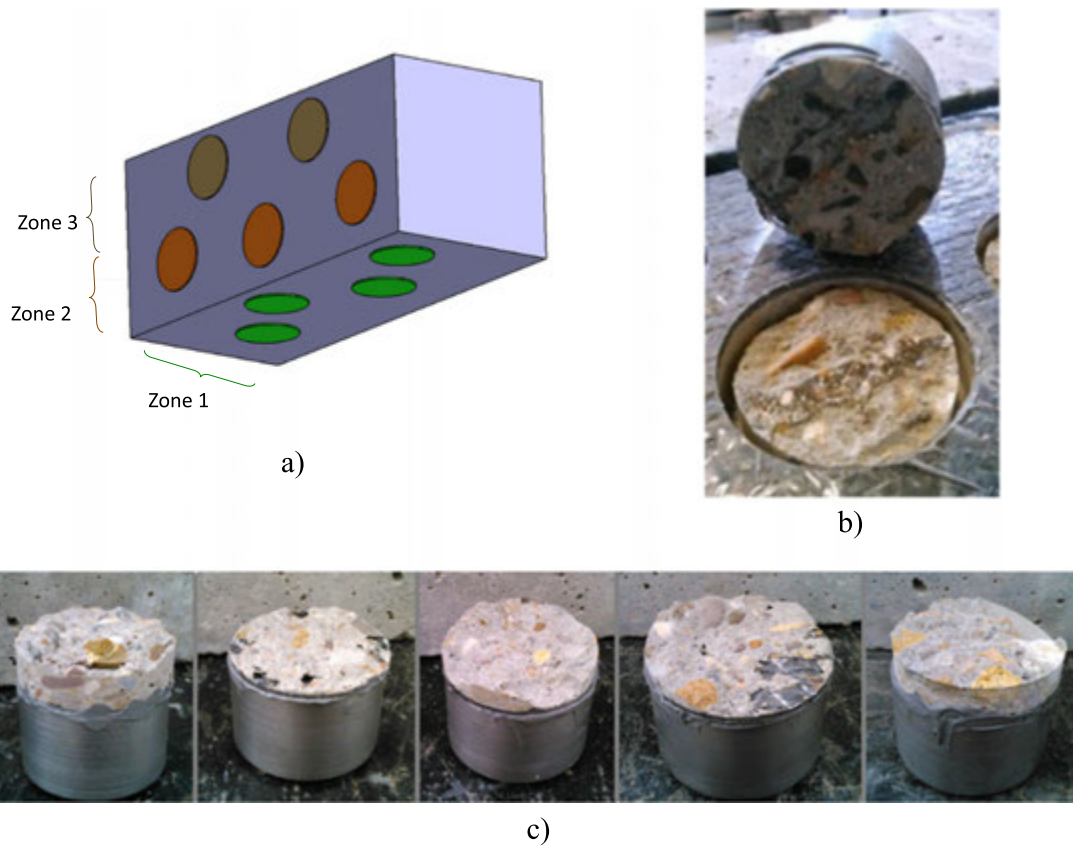


Fig. 9 (a) Locations in the concrete block of the trepanned samples, (b) detail of failure, and (c) some broken samples of the pull off test.

Table 2 Experimental results for the shear torsion tests

Surface treatment	Manufacturing procedure								
	Pre-cured			Hand lay-up			Infusion		
	Torque (Nm)	Std. Dev (Nm)	Cov (%)	Torque (Nm)	Std. Dev (Nm)	Cov (%)	Torque (Nm)	Std. Dev (Nm)	Cov (%)
Grinder	107 (6%)	7.90	7.38 ²	119 (18%)	10.50	8.82 ¹	111 (10%)	15.90	14.32 ²
Grit sandpaper	133 (32%)	11.23	8.44 ¹	101 (ref)	15.39	15.24 ²	136 (35%)	32.10	23.60 ¹



Fig. 10 Broken samples of the shear torsion test.

Test results for the shear torsion test are summarized in [Table 2](#). The lowest torque value (hand lay-up with grit sandpaper) is taken as the reference value (ref) with respect to which the rest of failure torque values are compared (in %) between brackets. The broken samples corresponding to grinder + infusion are shown in [Fig. 10](#). As done with the pull off tests, the superscripts in the covariance value indicate the location where the samples were obtained from the concrete block. For this test, again, all samples broke at the concrete and the differences in the torque value between the different manufacturing procedures and surface

treatments is much lower than that observed in the pull off test results. In this test, the fracture surface is not flat and there is presence of all concrete depths due to the characteristic helicoidal shape of the fracture surface under torque loadings.

Analysis of the Stress Singularities at the Sample's Corners

The trepanned sample introduced in Section "Problems Under Study" contains several points where the geometry and/or the material properties change abruptly. With the laminate bonded to the concrete block and the aluminum cylinder bonded to the laminate, at least there are 4 potential locations where a corner appears (taking also into account the single-material corner generated at the bottom surface of the trepanation in the concrete). These four single- and bi-material corners are the same for both test samples (pull off and shear torsion) and are depicted in Fig. 11.

From the top to the bottom, the corners are:

- (1) Corner 1: Bimaterial corner between the aluminum block and the adhesive.
- (2) Corner 2: Bimaterial corner between the adhesive and the carbon fiber laminate.
- (3) Corner 3: Bimaterial corner between the adhesive and the concrete block.
- (4) Corner 4: Single-material corner (concrete) at the bottom part of the trepanation.

The geometry of corners 1, 2 and 3 is equal, with two solid wedges of 90° with free-free boundary conditions and perfect adhesion at the bimaterial interface, while corner 4 has a single material free-free wedge of 270° . A simplified 2D generalized plane strain configuration of the corners will be considered.

As the aluminum, both adhesives and concrete block will be treated as isotropic materials. In this way, the results for the singularity analysis will be unique for the entire perimeter of the sample. Nevertheless, the carbon fiber laminate is orthotropic and at each point of the perimeter of the circumference the relative orientation of the fiber and the exterior surface normal changes. Taking 0° as the fiber direction (see Fig. 12), the corner configuration at different angles will be different as the mechanical properties of the laminate, measured with respect to a polar reference system centered at the cylinder center, change. In Fig. 12 three different angles (0° , 45° and 90°) are illustrated to clearly show how different the corner may be.

At 0° , the fiber is perpendicular to the free surface, while at 90° the fibers are parallel. For this particular corner configuration, the singularity results depend on the fiber orientation, although due to the symmetry of the problem, only a quarter part of the problem ($0^\circ < \text{angle} < 90^\circ$) needs to be calculated.

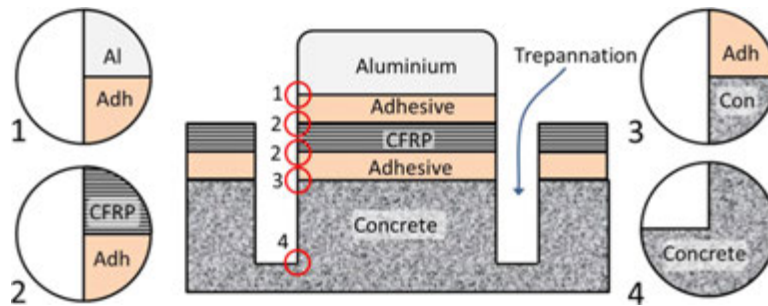


Fig. 11 Detail of the material corners appearing at the trepanned samples.

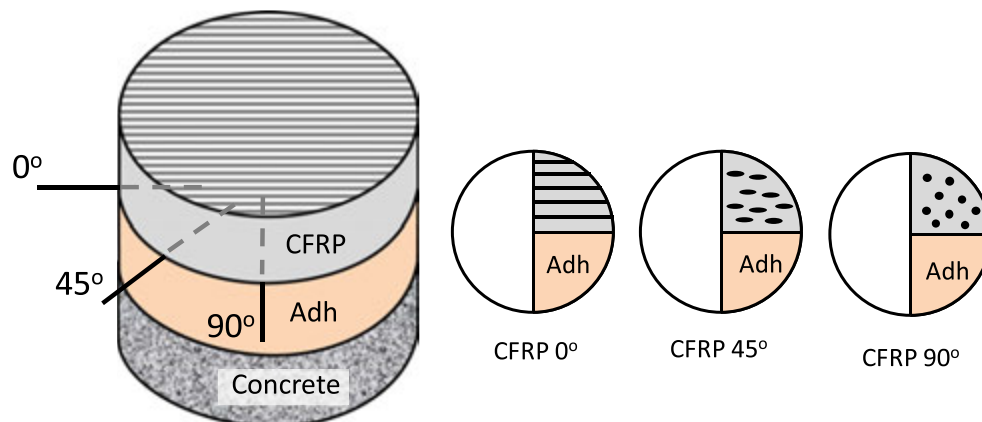
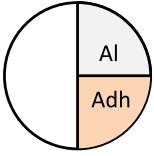
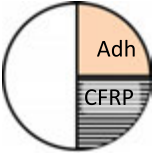
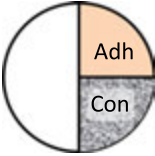
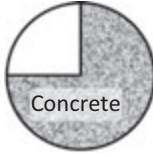
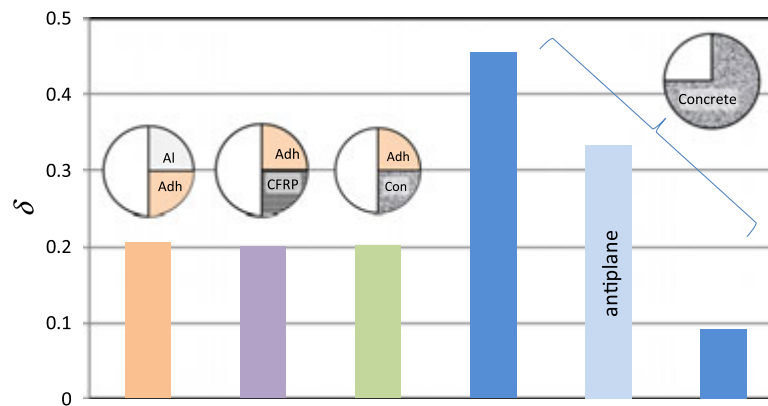


Fig. 12 Detail of the CFRP-Adhesive corner and its dependence with the fiber orientation.

Table 3 Orders of stress singularities in the material corners of the trepanned coupons

1	2	3	4
			
$\delta = 0.205393$	$\alpha = 0$ (and $\alpha = 90$) $\delta = 0.199914$ $\alpha = 15$ (and $\alpha = 75$) $\delta = 0.199852$ $\alpha = 30$ (and $\alpha = 60$) $\delta = 0.199733$ $\alpha = 45$ $\delta = 0.199675$	$\delta = 0.20065$	$\delta_1 = 0.455516$ (antiplane) $\delta_2 = 0.333333$ $\delta_3 = 0.0914708$

**Fig. 13** Orders of stress singularities in the material corners of the trepanned coupons.

The orders of the stress singularities, δ in Eq. (1), are summarized in Table 3 and illustrated in Fig. 13.

Configurations for $\alpha = 0^\circ, 15^\circ, 30^\circ, 45^\circ, 60^\circ, 75^\circ$ and 90° have been analyzed. The results are symmetric with respect to 45° and there are no significant variations in the values of the order of stress singularities for all fiber orientations calculated (differences in the fourth significant digit). For corners 1, 2 and 3 there is only one singularity term (only one order of stress singularity), while in the 270° concrete corner (corner 4) there are three orders of stress singularities (2 in plane and 1 antiplane).

It is noticeable that the three bimaterial corners have very similar values for the order of stress singularity (which can be considered as weak singularities, $\delta \approx 0.2$), while the single-material concrete corner has three singularity terms, the first in plane term being much severe ($\delta = 0.455$) than the other three corners, and the antiplane term ($\delta = 0.333$) also more severe than those of the other three corners.

The Generalized Stress Intensity Factors (K_k) in Eq. (1) depend on the applied load and the global geometry. The pull off test will predominantly activate the in-plane singularity terms, while the shear torsion test will predominantly activate the antiplane terms.

Discussion

All observed failures for both test configurations have shown a very brittle failure as all failures have occurred inside the concrete part of the specimens.

After the inspections of the fracture surfaces, it seems reasonable to admit that failure is not influenced by the singularity stress fields appearing at the material corners of the samples, as most of the samples broke at the concrete part, near, but not at the trepanation end, see Fig. 9(b).

For those samples where failure started at (or near) the bottom of the trepanation depth, singularity results support this type of failure, as the highest stress singularity value occurs at the concrete corner (at the bottom of the trepanation). But for the majority of the samples, the failure inside the concrete (but not starting at the corners) reveals that the weakest point of the sample, where the maximum stress reaches first the strength, is the bulk concrete, as illustrated.

This fact allows to conclude that, although it is not possible to obtain the adhesion strength of the composite to the concrete (because the failure did not involve this interface), this strength is higher than the characteristic strength of the concrete, and the bonding is successful.

Tests on Cantilever Coupons. Lap Shear and Lap Peeling Tests

Tests Description

A precured carbon fiber laminate is bonded for both tests (lap peeling and lap shear) to a concrete block with part of the laminate outside the concrete block. Two alternative surface preparations before bonding (grinder and grit sandpaper) were used, exactly in the same way as described previously in Section “Tests on Cylindrical Trepanned Coupons. Pull off and Shear Torsion Tests”.

The qualitative difference between both tests on cantilever coupons is basically the load direction, as introduced in Section “Materials and Methods”. For the lap shear test, the load is applied in the same plane of the laminate, creating mainly a shear stress distribution across the adhesive layer. For the lap peeling test, the load is applied perpendicularly to the composite laminate plane, creating, mainly, a peel stress distribution at the adhesive layer. In both tests the load application point is the laminate end outside the concrete block.

Both tests were carried out using an electromechanical testing machine Instron 4482. For the lap peeling test the samples were placed horizontally, whereas for the lap shear test, the samples were placed vertically. Fig. 14 shows the setup for the (1) lap peeling test and (2) lap shear test.

Coupons Preparation and Experimental Results

The dimensions of the concrete block (Fig. 15) are height $H = 150$ mm, width $H = 150$ mm and length $L_c = 600$ mm, while the composite laminate has $h = 2$ mm thickness and $w = 45$ mm width, and has a total length of $L = 400$ mm, 250 mm being inside the concrete block, from which the bonded length is $L_f = 200$ mm and $L_{free} = 50$ mm without being bonded.

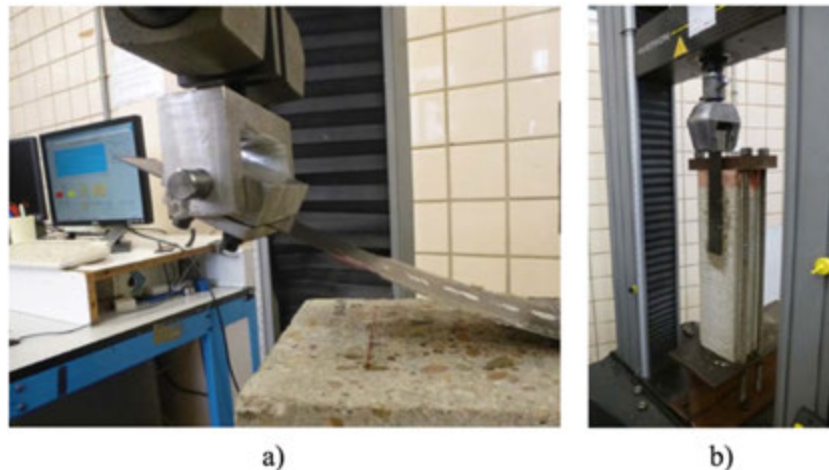


Fig. 14 Setup for (a) lap peeling test, (b) lap shear test.

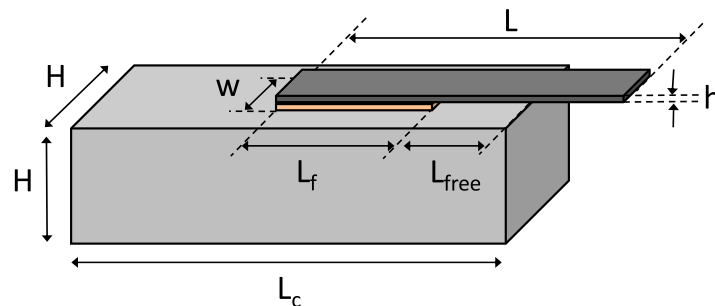


Fig. 15 Dimensions of the cantilever coupons.

Lap peeling tests may exhibit large displacements, thus, some precautions and special considerations have to be taken into account during the test. To avoid secondary bending moments appearing at the load point, a rotatory device grip was used (see Fig. 16). Also, the horizontal displacement of the concrete block was allowed to be free, by using some rollers below the concrete block. In fact, an intermediate piece with smooth surfaces, was placed between the concrete block and the rolls to avoid the roughness of the concrete surface to inhibit the rolling movement. Due to the fact that the own weight of the concrete block (317 N) is higher than the characteristic failure load initiation for this peeling test (peak values around 100 N for grinder treated samples, and below 80 N for grit sandpaper treated samples), no additional device was considered necessary to avoid the vertical displacement of the whole sample. For the lap shear tests, failure load is much greater (between 20.000 N and 25.000 N), thus, the concrete block was fixed to the lowest part of the testing machine using a metallic frame.

Test results for the lap peeling tests, with the two surface treatments, are shown in Fig. 17. Five samples were tested with each surface treatment. The displacement was measured with the cross-head of the testing machine. Two main differences can be observed between the two set of samples: (1) after an initial linear elastic behavior in both cases, grinder surface treatment samples show a higher initial failure load and lower dispersion (see Table 4), and (2) after the first initial failure, grinder samples show slight load increments after each failure step.

These results are reasonably easy to understand with the observation of the failure surfaces, see Fig. 18(a) grinder and b) grit sandpaper, showing failure surfaces in the lap peeling tests. It is clear that, for grinder samples, the failure has progressed mainly inside the concrete block, while in the grit sandpaper samples, the failure appears mainly at the adhesive-concrete interface, on the concrete side. It is worth to note that evidences of lack of bonding between concrete and composite have been observed in some areas of the grinder samples, see Fig. 18(c). These imperfections are mainly due to the unavoidable irregularities of the

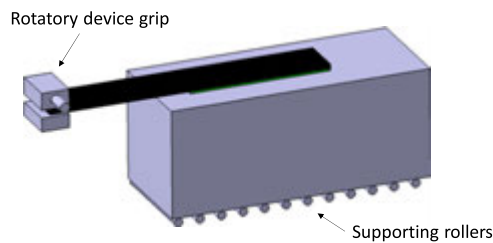


Fig. 16 Scheme of the test set-up of the lap peeling test.

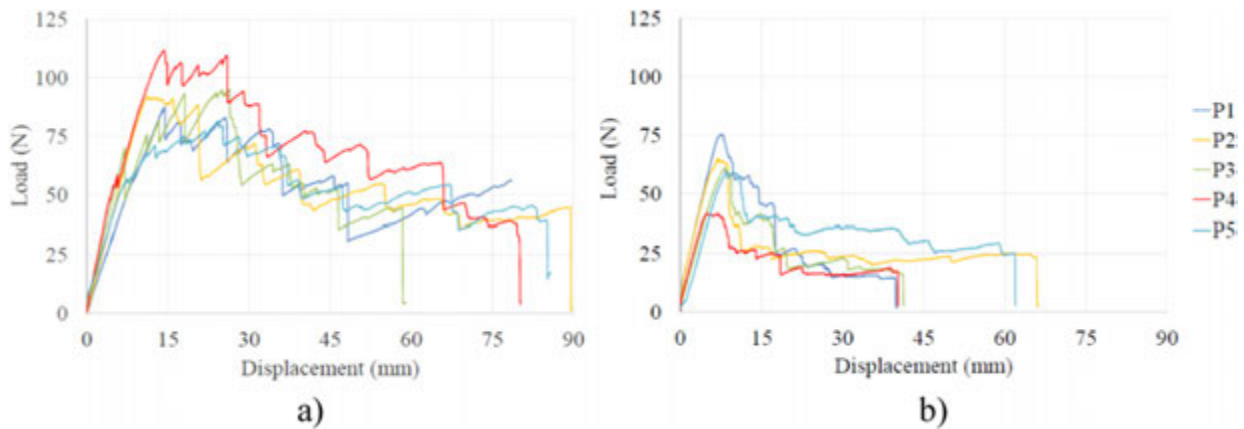


Fig. 17 Load-displacement curves for the lap peeling tests with (a) grinder, and (b) grit sandpaper surface treatments.

Table 4 Results for lap peeling tests with (a) grinder, and (b) grit sandpaper surface treatments

Surface treatment	Mean max load (N)	Std. Dev. (N)	Covariance (%)
Grinder	93.71	11.34	12.10
Grit sandpaper	60.70	12.29	20.25

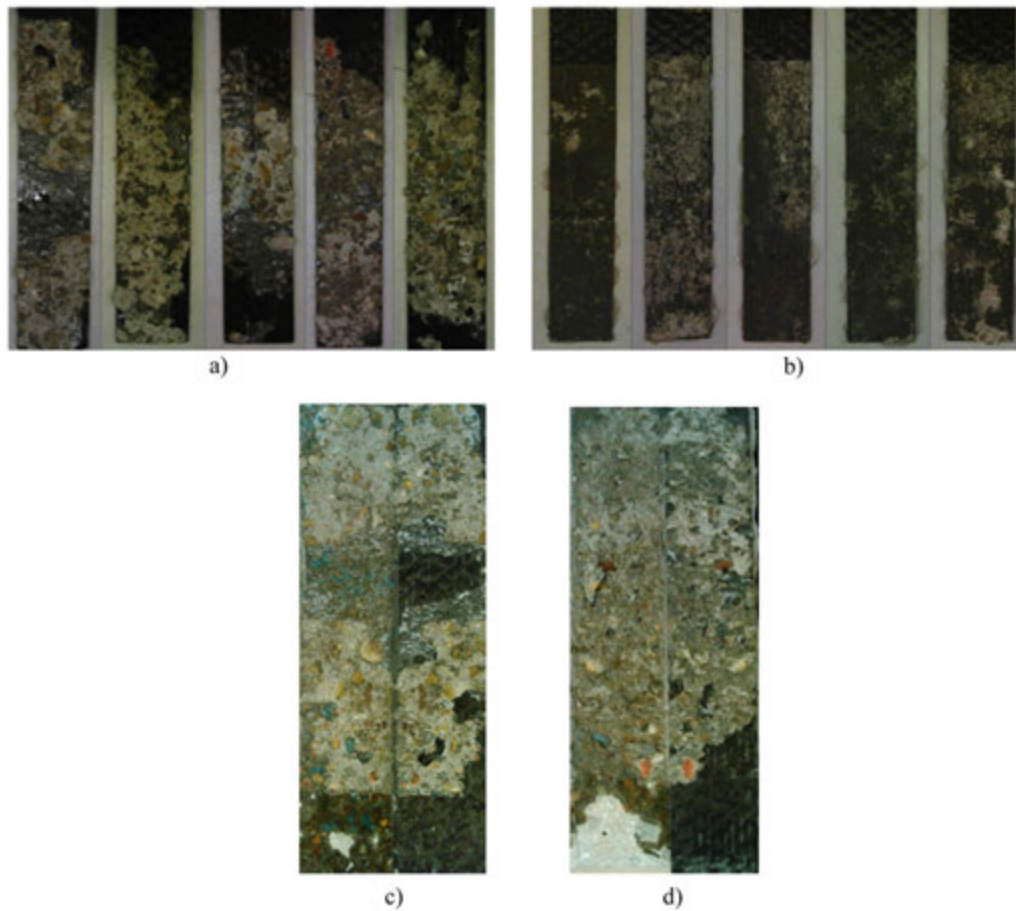


Fig. 18 Failure surfaces in the lap peeling tests: (a) for grinder samples, (b) for grit sandpaper samples, (c) detail of failure with lack of bonding in a grinder sample, and (d) detail of failure without imperfections in a grinder sample.

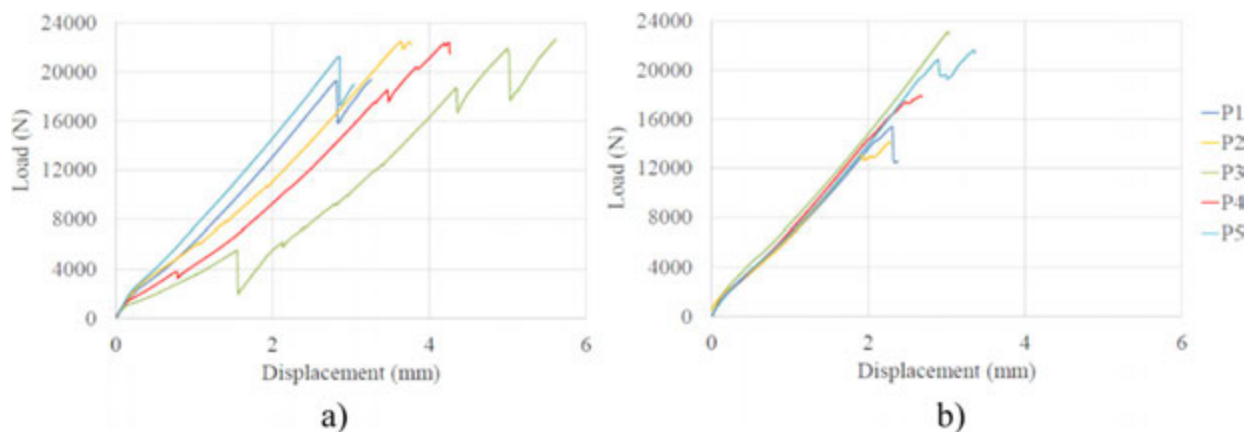


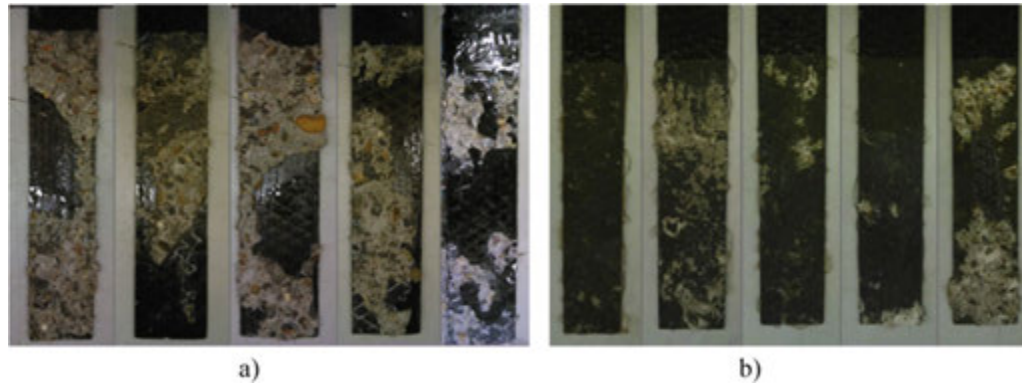
Fig. 19 Load-displacement curves for the lap shear tests with (a) grinder, and (b) grit sandpaper surface treatments.

concrete surface, which prevents the flat precured composite to follow satisfactorily the concrete surface. **Fig. 18(d)**, nevertheless, shows a failure surface, where this type of imperfection does not appear, a failure inside the concrete being observed.

Results for the lap shear tests, with the two surface treatments, are shown in **Fig. 19**. In this case, a set of five samples were tested with each surface treatment also. Now the difference for the maximum load is less significant than in the previous case, although again, the grinder surface treatment samples show slightly higher failure loads and less dispersion (see **Table 5**). Some peaks at small loads that appear in the grinder coupons, related to previous failures until reaching the peak, can be appreciated. For lap shear tests, the failure behavior is more brittle than for the lap peeling tests.

Table 5 Results for lap shear tests with a) grinder, and b) grit sandpaper surface treatments

Surface treatment	Mean max load (N)	Stad. Dev. (N)	Covariance (%)
Grinder	21,631	1392	6.44
Grit sandpaper	18,431	3873	21.02

**Fig. 20** Failure surfaces in the lap shear tests for (a) grinder, and (b) grit sandpaper samples.**Table 6** Geometric dimensions of the beam models and FEM computations, see Fig. 16.

	Nominal value	Adjusted value
w (laminate width)	45 mm	45 mm
h (laminate thickness)	2.0 mm	1.7 mm
L_c (concrete block length)	600 mm	600 mm
H (concrete block height and width)	150 mm	150 mm
t (adhesive film thickness)	0.1 mm	0.1 mm
$a = L_c - L_f$ (crack length)	75 mm	82 mm

Pictures of the fracture surfaces are shown in Fig. 20 for both set of samples (grinder and grit sandpaper). It is clear that the presence of concrete can be observed in a more pronounced way in the grinder samples than in the grit sandpaper samples.

Analytical Beam Models

Two beam models, one for lap peeling and another for the lap shear test, have been developed from which the evolution of stress and displacement fields during the load process can be analyzed. They will allow experimental measurements to be understood and also the characteristics of more sophisticated models based on FEM to be adjusted. Two main characteristics have to be considered for the adjustment of the predictions: the strength and the toughness of the joint.

The basic geometry was shown in Fig. 16, and the main nominal dimensions and values of the properties of the materials are summarized in the Nominal value columns of Tables 6 and 7 respectively ("Adjusted value" columns will be introduced later on).

Analytical model for the lap peeling test

As has been described above, the manufacturing process employed for the composite laminate of these coupons is completely manual and it leads to a high variability, inherent to the process, in the final properties obtained, usually lower than the nominal ones. To obtain an estimation of the actual properties, a cantilever beam model has been employed. The displacement at the tip of the composite laminate, v , versus the force applied at the tip of the beam, F , is:

$$v = 4 \frac{F a^3}{E w h^3} \quad (3)$$

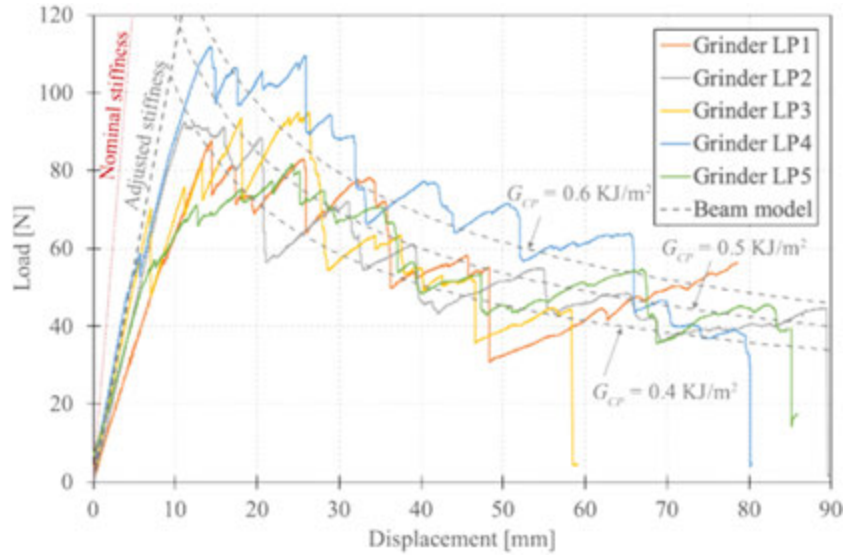
where w is the width of the section, h is the height of the section, a is the length of the beam (crack length) and E is the elastic modulus of the composite in the direction of the fibers (E_{11}).

With this model, the initial slope of the F - v curve can be estimated. More sophisticated models which contemplate geometrical non-linearities could also be employed, see for instance (Williams, 1987; Sundararaman and Davidson, 1997).

Table 7 Material properties. ⁽¹⁾ values obtained from Eurocode 2: $E_c = 8500 f_{cm}^{1/3}$, $f_{cm} = f_{ck} + 8$ MPa.

	Nominal value	Adjusted value
E_{11} (composite laminate)	154000 MPa	111000 MPa
E_{22} ($= E_{33}$) (composite laminate)	8500 MPa	8500 MPa
ν_{12} ($= \nu_{13}$) (composite laminate)	0.35	0.35
G_{12} ($= G_{13}$) (composite laminate)	4200 MPa	4200 MPa
E_c (concrete)	29779 MPa ⁽¹⁾	29779 MPa
ν_c (concrete)	0.1875 ⁽¹⁾	0.1875
E_a (adhesive)	270 MPa	270 MPa
G_a (adhesive)	114 MPa	114 MPa

Note: Eurocode 2, 2004. N 1992-1-1: Eurocode 2: Design of Concrete Structures – Part 1-1: General Rules and Rules for Buildings. Brussels: CEN.

**Fig. 21** Comparison of grinder lap peeling measurements and analytical beam results.

In order to model the evolution of the load when the crack propagates during the test, the energy release rate (ERR) concept has been used. If the value of ERR reaches the critical value G_{cr} the crack progresses. This fact occurs when the load and the displacement, which are related by Eq. (3), achieve a critical value, F_{cr} and v_{cr} , obtained from the definition of G :

$$G = \frac{1}{w} \left(\frac{\partial W}{\partial a} - \frac{\partial U}{\partial a} \right) \quad (4)$$

$dW = F dv$ is the variation in the external work and $dU = \int_0^a M \kappa dx$ is the variation in the internal energy, M and κ being the bending moment and the curvature respectively.

Crack grows when G reaches a critical value, that depends on the mode mixity. Calling G_{CP} the critical value for the lap peeling test and taking $G = G_{CP}$, a relation between the load, F_{cr} and the displacement, v_{cr} , for the crack to progress is obtained from Eq. (4). Then, taking into consideration that F and v in the model are related by Eq. (3), these critical values are easily obtained:

$$F_{cr} = w \sqrt{\frac{1}{6} E G_{CP} h \frac{h^2}{a^2}} \quad v_{cr} = \sqrt{\frac{8 G_{CP} a^3}{3 E h^3}} \quad (5)$$

A comparison between the experimental data and the evolution obtained with this beam model for 3 selected values of G_{CP} are shown in Fig. 21 for the grinder treated coupons. Two considerations deserve to be mentioned: on the one hand the initial linear evolution, prior to the crack growth, and on the other hand, the influence on the load-displacement curve of the crack progression.

The red dotted line represents the stiffness obtained with the nominal values presented in the first column in Table 7. It can be seen that this evolution is stiffer than that obtained experimentally. In order to understand and justify the main causes and to adjust the actual stiffness of the coupon, the parameters that appear in Eq. (3) have been reconsidered, taking into consideration the aforementioned influence of the manufacturing process employed.

- (1) E_{11} : stiffness of the laminate in the longitudinal direction. The lay-up technique can produce misalignment in the fibers that can justify a reduction of this property even, based on our experience in testing composites, up to a 30% ($107.8 \text{ GPa} \leq E_{11} \leq 154 \text{ GPa}$).

- (2) w : width of the laminate. The cut of the coupons was made with a precision diamond disc saw, then considerable deviations in this parameter ($w = 45$ mm) are not expected.
- (3) h : thickness of the laminate. Even having used a vacuum bag compaction in addition to the manual compaction, a non uniform fiber distribution through the thickness is obtained, a thin layer rich in resin being formed over the laminate. It justifies a decrease in the nominal thickness up to a 20% ($1.6 \text{ mm} \leq h \leq 2 \text{ mm}$).
- (4) a : crack length, which in the model equals the distance from the load application point to the position of the crack tip, i.e., the length of the bar in the analytical beam model. The application of the adhesive was made carefully, but it is inevitable (even with the use of an anti-adherent layer) to obtain uncertainties in this measurement at the beginning and during the test.
- (5) In addition to the parameters described before, taking into account that the value of the displacement given by the testing machine refers to the movement of the grip, the initial stiffness can be affected by the own strains of the gripping fixture. Nevertheless, due to the nature of the peeling test, it is reasonable to suppose that the test fixture is stiff enough (compared with that of the specimen) to neglect its influence.

After several attempts, a family of values that adjust the experimental results has been obtained (see gray dashed line in Fig. 17). These values are presented in column "Adjusted values" of Tables 6 and 7.

Fig. 17 also shows the F - v evolution when the crack propagates, for three values of G_c : 0.4, 0.5 and 0.6 kJ/m², that bound the experimental measurements and represent correctly the evolution of the tests.

Fig. 22 shows the same comparison for the grit sandpaper treated coupons. Axis scales have been maintained for comparison purpose. The initial stiffness coincides with that obtained with the grinder treated coupons, corroborating the previous reasonings. However, the toughness in this case is near 5 times smaller than that obtained previously.

The analysis carried out has proved that a simple beam model can be used to adjust both the initial stiffness of the coupons and the toughness of the joint, that initially could be considered near mode I, but nothing can be concluded about its strength. More sophisticated numerical models are needed for that and also in order to verify mixity.

Analytical model for the lap shear test

For the lap shear tests, another beam model has been developed, with the same adjusted geometrical dimensions and properties of the previous model, except for the beam length, that is 200 mm now. In this case, a tensile load is applied in the longitudinal direction, the relation between the displacement of the laminate tip in the direction of the load, u , and the load applied, F , is:

$$u = \frac{F a}{E w h} \quad (6)$$

In this model, in order to apply expression (4), $dW = F du$ and $dU = \int_0^a N d\epsilon dx$, N being the axial internal force and ϵ the elongation per unit length. As has been done with the lap peeling beam model, crack grows when G reaches a critical value that, now, it is denoted G_{CS} . Taking $G = G_{CS}$ in Eq. (4), and considering the relation defined by Eq. (6), the following critical values are obtained:

$$F_{cr} = w \sqrt{2 G_{CS} E h} \quad u_{cr} = \sqrt{2 \frac{G_{CS} h a}{E h}} \quad (7)$$

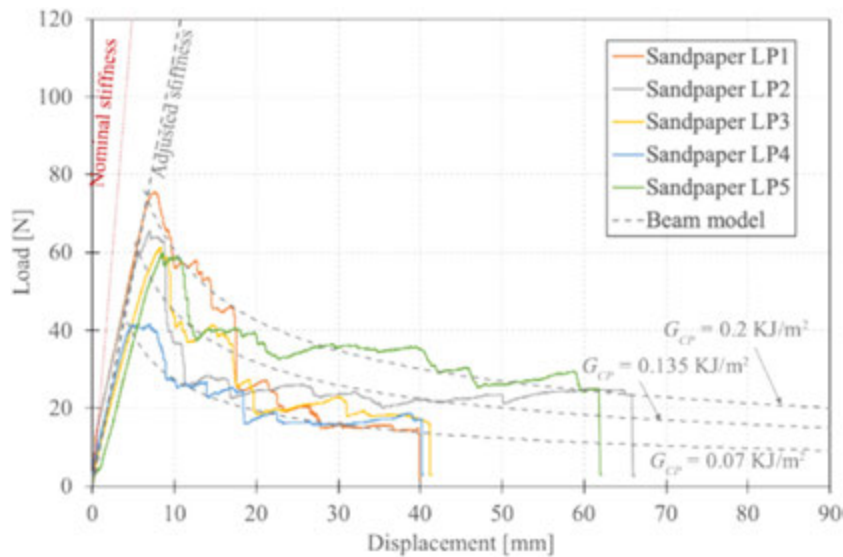


Fig. 22 Comparison of grit sandpaper lap peeling measurements and analytical beam results.

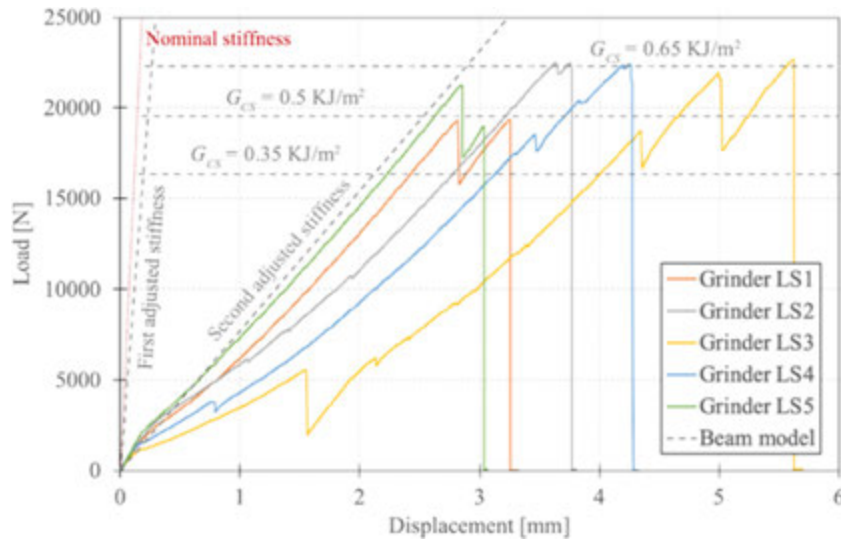


Fig. 23 Comparison of grinder lap shear measurements and analytical beam results.

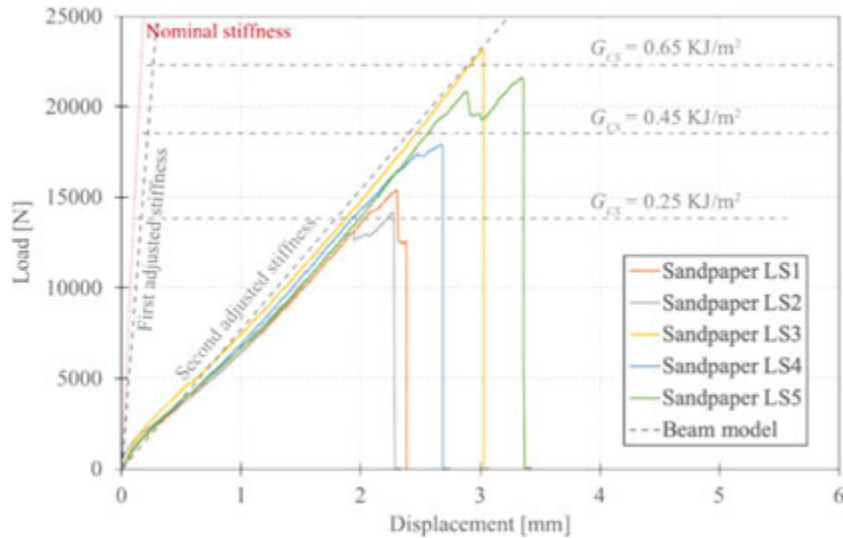


Fig. 24 Comparison of grit sandpaper lap peeling measurements and analytical beam results.

It is noticeable that now the critical force does not depend on the crack length.

Figs. 23 and 24 show the experimental results for both grinder and grit sandpaper treated coupons, respectively, and the results of the beam tensile model, for three values of the toughness, G_{CS} , for each group of coupons.

It can be seen that, again, the stiffness of the model is higher than that obtained in the experiments, which are similar in both family of coupons, even when the adjusted values of Tables 6 and 7 are used. Loads are much higher now than in the lap peeling test, thus, this difference has been attributed to adjustment processes that occur between the coupon and the grip and inside the gripping system itself, where the displacement is measured. The main cause is the adjustment and elastic string that appear in the mechanical joints of the gripping system during the application of the load. Differences of this kind are commonly observed, in accordance with our experience, in the tensile test of unidirectional composite coupons. This initial stiffness could be adjusted increasing artificially the length of the beam model up to a modified length of $\hat{a} = 1100$ mm or, equivalently and with a higher physical sense, by introducing a spring of elastic constant K_m . A value of $K_m = 8491$ N/mm has been taken for a spring located between the load application point and the tip of the laminate, which has been found to produce a satisfactory agreement between analytical predictions and experimental measurements before the crack grows. This is referred in Figs. 23 and 24 as “Second adjusted stiffness” curves.

When the crack is growing, predicted analytical evolutions follow horizontal straight lines, as shown by Eq. (7), but no evidence of this behavior is noticed in the tests. It is reasonable to assume that the lack of agreement between the analytical predictions and experimental measurements might be due to the simplicity of the beam model used. An additional

complementary explanation could be that the length of the specimens is not long enough for the crack to progress according to the hypotheses considered.

However, assuming that the critical value is defined by the top value, the values of G_C that adjust the beam model results to the experimental measurements are similar for both families of coupons, a higher dispersion appearing in the grit sandpaper treated coupons.

It seems that it can be assumed that the crack grows under mode II conditions, but in order to verify this, and to know the distribution of stress along the interface and the effect of the strength of the joint, a corresponding finite element model has been developed.

FEM Models

Finite element simulations have been performed in order to obtain specific details about the evolution of the stress and displacement fields. The software used has been Abaqus® 2019. Several 2D plane stress models have been developed considering a unitary width.

Geometrical non-linearities have been taken into account. A thin layer (0.01 mm) of cohesive elements has been used to model the joint between the concrete block and the composite laminate, following a traction-separation uncoupled law, with the characteristics shown in [Table 7](#), and a linear degradation evolution, ([Reinoso et al., 2016](#)).

Adjustments performed with the beam models have been taken as a guide in order to define all features of the model. In particular, for the interface, G_{IC} values have been taken from the adjustment performed between the analytical model and lap peeling test measurements, and similarly for G_{IIC} with respect to lap shear tests. Thus, $G_{IC} = G_{IIC} = 0.5 \text{ kJ/m}^2$ are used to simulate the grinder coupons group, and $G_{IC} = 0.135 \text{ kJ/m}^2$ and $G_{IIC} = 0.5 \text{ kJ/m}^2$ are used for the sandpaper group. The values corresponding to the grinder coupons group can be considered a very strange combination, because G_{IIC} is usually higher (two or three times, as happens with the sandpaper group, or even more) than G_{IC} . In any case, these values have been taken just as initial values for a parametric study.

Benzeggagh-Kenane mixity model, [Benzeggagh and Kenane \(1996\)](#), has been used, having taken η parameter equal to 2.5, [Eq. \(2\)](#).

Other parameters are needed for the cohesive law, which are the tensile and the shear strengths. No experimental values are available, but some considerations can help to quote them.

First, in order to obtain a coherent cohesive law, strength must fulfill the following conditions, [Távora et al. \(2019\)](#):

$$\begin{aligned} S_N &\leq \sqrt{\frac{2G_{IC}E}{t}} = 52.0 \text{ MPa} \\ S_S &\leq \sqrt{\frac{2G_{IIC}G}{t}} = 33.8 \text{ MPa} \end{aligned} \quad (8)$$

In fact, in order to avoid convergence difficulties, $\leq$ sign should be replaced by $\ll$. In the other limit ($=$ sign), the cohesive law become a linear elastic brittle interface model (LEBIM, [Mantič et al., 2015](#)) that, although being simpler than the cohesive law, presents many convergence problems if the standard solution procedure of Abaqus is used. Thus, conditions in [Eq. \(8\)](#) have to be considered in the form: $S_N \ll 52 \text{ MPa}$ and $S_S \ll 33.8 \text{ MPa}$. Notice that these values are of the order of the nominal tensile and shear strength of the resin (about 20 and 30 MPa respectively).

Second, visual inspections of the broken interface evidence that fractures were adhesive, which means that crack grows through a superficial concrete layer, and not inside the adhesive. Thus, it is more realistic to use the tensile and shear strength of the concrete. [Eurocode 2 \(2004\)](#) proposes a formula to estimate the tensile strength of a concrete from its characteristic compressive strength, f_{ck} . Coupons are manufactured with a concrete of 35 MPa, the tensile strength resulting: $S_T = 0.3 f_{ck}^{2/3} = 3.21 \text{ MPa}$. For the shear strength the standard assumes that the shear strength equals the tensile strength, but if a joint is considered, the shear strength become smaller, it being of an order of 1.13 MPa for a very rough joint, for example, the grinder coupons group, or of an order of 0.565 MPa for the grit sandpaper coupon group.

Another important issue from the point of view of the convergence of the cohesive model is the number of elements in the resulting cohesive zone. In this sense, it is usual to recommend having at least 10 elements in it. There are several proposals that estimate the cohesive zone length from the stiffness of the adherent, E , the toughness, G_C , and the strength, S , the [Hillerborg et al., \(1976\)](#) and [Rice \(1980\)](#) model being those most commonly used in the literature. Assuming $E = 8500 \text{ MPa}$, $G_C = 0.5 \text{ kJ/m}^2$, and $S = 10 \text{ MPa}$, the values obtained are 42.5 mm and 37.6 mm respectively. Thus, it would be sufficient to use 3 mm length elements along the interface. However, this can also be considered an initial estimation because the problems are not the same, and if the use of artificial tools to facilitate convergence (for example, viscosity) are wanted to be avoided, a smaller size would be required. After several attempts, the element along the interface in the mesh used for all the finite element simulation is that corresponding to 0.5 mm length.

Lap peeling test

For the lap peeling test, a displacement has been applied at the end of the composite laminate. The grip has been modeled by means of a multipoint constraint at a point placed 8 mm high and 15 mm length from the tip of the composite laminate. Due to

the fact that the concrete block was not separated from the ground in the laboratory tests, the weight of the concrete has not been considered and a displacement condition has been imposed at the inferior face of the concrete block.

FEM results are shown in Fig. 25 with the experimental measurements and the analytical prediction. Two FEM models appear, referred by 4 numbers that represent: G_{IC} , G_{IIC} , S_T and S_S respectively.

The initial stiffness of the FEM model is slightly smaller than that obtained from the analytical model. This is because the foundation in the analytical beam model is rigid, whereas that of the FEM model is elastic. However, the FEM stiffness belongs to the range obtained experimentally.

Evolutions of both FEM solutions are similar each other and evolve between the experimental measurements. However, they do not exactly coincide, which, considering that the unique difference is the S_S value, may indicate that the process does not correspond to a pure mode I, see Cañas *et al.*, (2018) for a discussion about the mixity in peeling tests.

Results for the sandpaper coupon group are shown in Fig. 26. where four FEM simulation are included. The effect of the reduction of G_{IC} is evident if compared with results in Fig. 16, but conclusions are similar when focused on FEM (0.135, 0.5, 3.21, 0.565) and FEM (0.135, 0.5, 3.21, 3.21) curves. Besides, it is noticeable that FEM (0.135, 0.5, 3.21, 0.565), FEM (0.135, 0.5, 6.5, 6.5) and FEM (0.135, 0.5, 10., 10.) curves are indistinguishable, which means that strength has no influence (if ratio S_T/S_N remains the same).

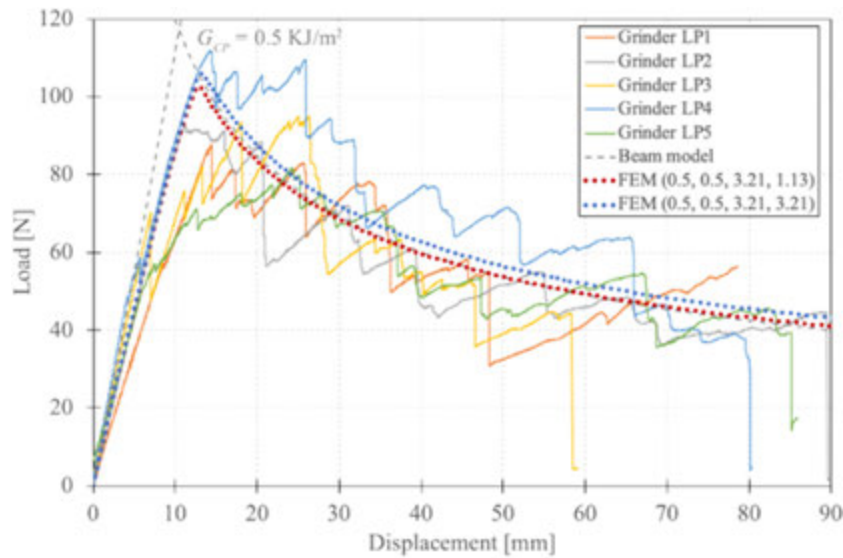


Fig. 25 Comparison of grinder lap peeling measurements with FEM results.

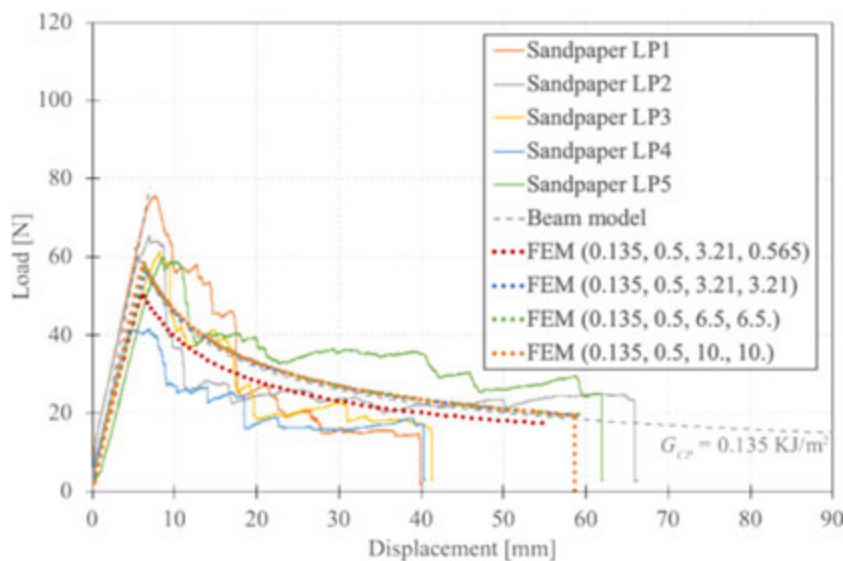


Fig. 26 Comparison of grit sandpaper lap peeling measurements with FEM results.

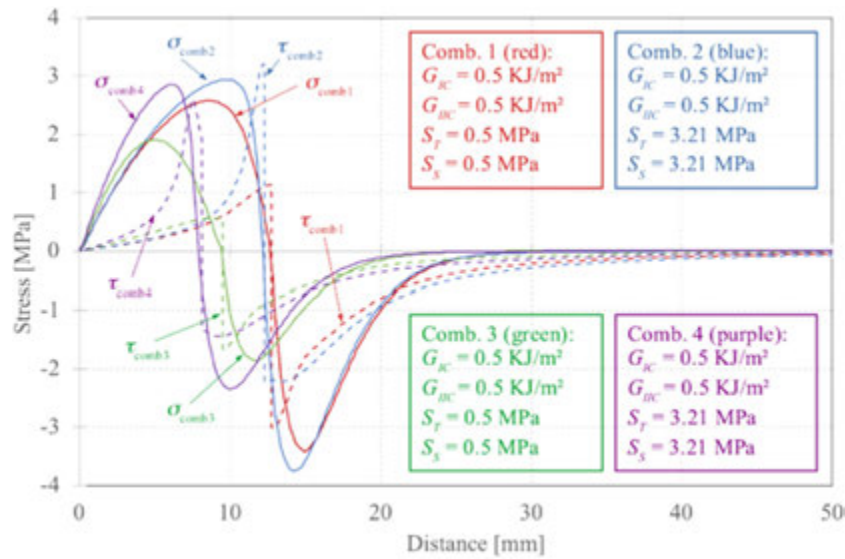


Fig. 27 Normal and shear traction distribution along the interface for four combinations of lap peeling test.

Fig. 27 shows the normal (continuous lines) and shear (dashed lines) traction distributions for four of the combinations considered (red, green, blue and purple in the figure) versus the distance to the crack tip. It is worth to be mentioned that the shape of the curves remains the same for each combination, translating with the crack tip.

It is clearly noticed the presence of not negligible shear stresses that confirm the mixicity of the crack tip conditions. The length of the cohesive zone, which approximately corresponds to the distance from the crack tip ($x = 0$) to the maximum of the tensile stress curve (which have resulted smaller than predicted by Hillerborg *et al.*, (1976) or Rice (1980)), is greater if G_{IC} increases (green and purple versus red and blue curves). It is also worth to be noticed that the shear stresses are relatively small along the cohesive zone, the maximum tangential stress being outside this zone.

Those curves also show that, it being predominantly a mode I test, the greater the shear strength, the greater the stress (normal and tangential) levels (purple versus green, and blue versus red).

Lap shear test

In the finite element model for lap shear tests, a spring with stiffness constant equals 8700 N/mm, as adjusted by the analytical study, has been included in order to model the effect of the stiffness of the gripping system. Although grinder and sandpaper experimental results are very similar, results are presented separately for both groups.

Now several FEM model configurations are considered, with $S_S = 0.565, 1.13, 3.21$ and 6.5 MPa, G_{IC} being 0.135 or 0.5 kJ/m² and G_{IIC} equals 0.5 kJ/m². Results are presented in **Figs. 28** and **29**, with those of the experimental tests and of the analytical beam model. The same meaning as before has been adopted for referencing each FEM configuration: (G_{IC} , G_{IIC} , S_T , S_S).

FEM predictions are very similar with those from the analytical beam evolution, but they do not reach the critical load defined from the assumed G_C . This fact is especially noticeable for those configurations with small S_S (0.565 and 1.13 MPa) and it justifies the brittle fracture nature found in these tests. Only for configurations with $S_S = 6.5$ MPa, a small plateau has been obtained, that providing the system some ductility.

In order to understand the phenomenon, the normal (continuous lines) and shear (dashed lines) traction distributions for a specific configuration: $G_{IC} = 0.135$ kJ/m², $G_{IIC} = 0.5$ kJ/m², $S_T = 3.21$ MPa, $S_S = 3.21$ MPa, are presented in **Fig. 30** for three specific load values during the test.

The first instant (in red) corresponds to a displacement of the grip of 2 mm, which in turns corresponds to a load of $F_1 = 14.3$ kN. Approximately half of the interface belongs to the cohesive zone, that starts at the crack tip (at $x = 0$) and finishes at the point where τ reaches the maximum value that coincides with the shear strength (3.21 MPa). Normal stresses are relatively small, presenting a concentration at the tip of the crack.

The second instant (in blue) corresponds with the point at which a cohesive zone is full developed. This point coincides with the maximum load carried out by the model ($F_2 = 17.99$ kN). Notice that most of the interface length is degraded by the cohesive model, and normal stresses are almost null along the full interface.

The third instant (in green) is when the full interface started to downgrade, load being slightly smaller than the maximum, $F_3 = 16.07$ kN. Notice that the crack has grown only a little length from the distribution for F_2 (in blue).

Hereinafter, all the points along the interface are damaged in some grade (shear traction and relative tangential displacement of each point are following the descendent portion of the cohesive law shown in **Fig. 6**), what means that shear force transferred throughout the interface can neither be increased nor maintained. However, load has to remain constant for the crack to grow

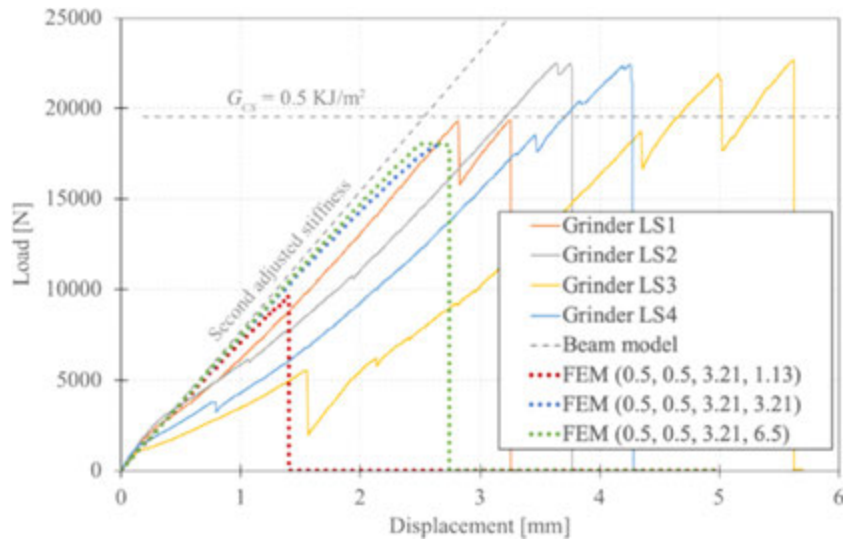


Fig. 28 Comparison of grinder lap shear measurements with FEM results.

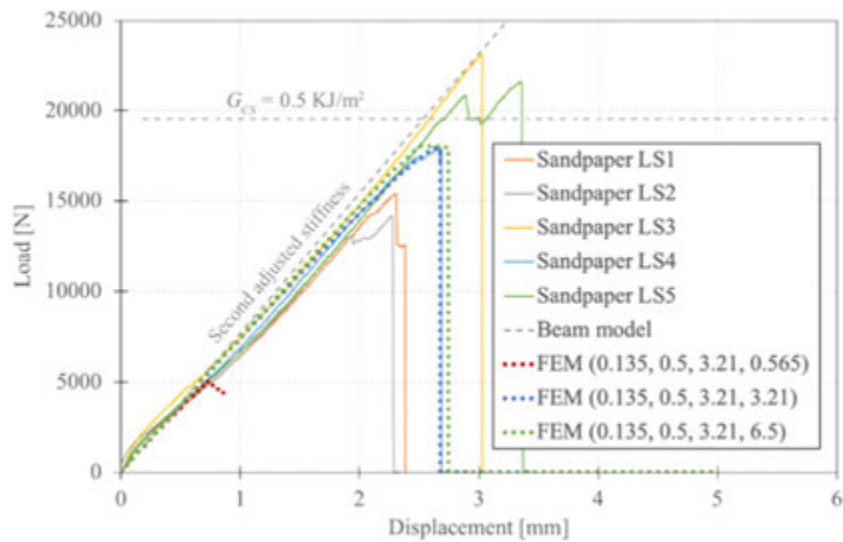


Fig. 29 Comparison of grit sandpaper lap shear measurements with FEM results.

(as predicted by Eq. (7) of the analytical beam model). Both conditions imply that the crack growth becomes unstable and they justify the sudden breakdown of all lap shear experimental tests and the convergence problems arising in the numerical model.

Discussion

Due to the fact that failures for all the coupons occur through the interface between the concrete and the adhesive or inside the concrete, rupture can be considered cohesive, the process being controlled by the strength and the toughness of the concrete.

Experimental measurements had been obtained directly from the grip system. A preliminary analysis using beam models has proved the influence of many sources of imperfections on the predicted evolutions. Taking into account these considerations, nominal characteristics of the coupons have been modified, the experimental results being properly adjusted. These adjusted properties have been considered for the FE models. The progression of the crack in these models has been taken into account using a small thickness layer of cohesive elements, with a traction-separation law and a linear degradation.

Some conclusions can be deduced from FEM results.

The behavior of the lap peeling test has been found to be reasonably progressive. The traction distributions shown in Fig. 27 have proved that conditions do not correspond to a pure mode I of fracture because non null shear stresses are found at the

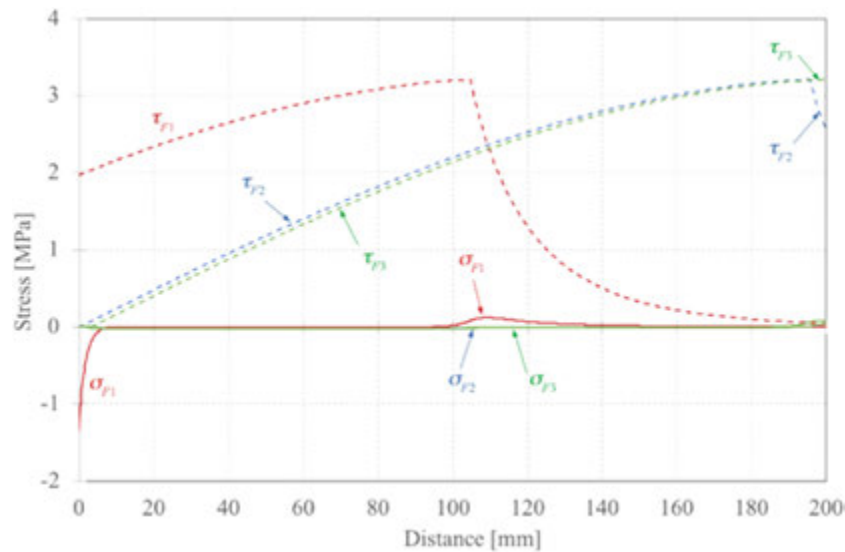


Fig. 30 Normal and shear traction distribution along the interface for three specific load values: $F_1 = 14.3$ kN, $F_2 = 17.99$ kN and $F_3 = 16.07$ kN in the lap shear test.

cohesive zone in the crack tip. However, satisfactory results have been obtained using FEM models if G_{IC} is assumed to be equal to G_{CP} , thus G_{CP} provides a reasonably value for the G_{IC} of the joint. On the contrary, the models have not allowed the strength of the joint to be validated.

On the contrary, lap shear tests have shown a brittle behavior, as a consequence of the simultaneous break of all the interface. FEM analysis indicates that conditions at the crack tip are very near to pure mode II, but the length of the specimen had been insufficient to notice the plateau value predicted by the analytical beam and the FEM models, thus, G_{IIC} cannot be obtained. However, an adjustment of the FEM results to the experimental measurements, as has been shown in Figs. 28 and 29, allows the shear strength of the cohesive zone model to be estimated.

In conclusions, and for the configurations considered in this work, on the one hand lap peeling tests allowed the G_{IC} to be measured, it being 0.5 kJ/m^2 for the grinder coupons and 0.135 kJ/m^2 for the sandpaper coupons, but no estimation of the tension strength could be obtained. And, on the other hand, lap shear tests allowed an estimation of the shear strength (for a cohesive zone model), S_s , to be obtained, it being 3.21 MPa for both grinder and sandpaper coupons, but G_{IIC} could not be measured because of the length of the specimen.

Conclusions

A general study on joining polymer matrix composites to concrete for structural applications has been carried out. Four tests (pull off, shear torsion, lap shear and lap peeling tests), involving different manufacturing procedures and surface preparations have been performed and stress singularity and numerical analyses have been developed to understand and complement the experimental results obtained.

The tests have allowed several properties related to the quality of the composite-concrete joint, as the peeling failure load, the flatwise tensile failure load and its lap shear failure load, to be studied.

From the tests on trepanned coupons, it has been seen that the manufacturing technique does not significantly affect the performance of the coupons, as the failure has occurred at the concrete. The tests on cantilever coupons have revealed that the pre-cured manufacturing technique is not desirable, as impedes the composite to follow the irregularities of the concrete, leaving zones rich in resin, where an intimate contact between adherents does not exist. Thus, the infusion process is recommended, due also to its inherent possibility of automation.

Regarding the surface treatments, the grinder is recommended, as entails higher properties for the joint, as seen in the tests on cantilever coupons. Again, no conclusions in this sense have been obtained in the tests on trepanned coupons, as the failure occurred at the concrete.

Once the experimental results were available, stress singularity analyses and numerical simulations were carried out to support the representativity of the results found. In the case of pull off and shear torsion tests, the main concern was the existence of material corners that could be influencing the performance of the joint. These materials corners involve singular stress states that must be properly modeled to get the parameters that control the potential appearance of the failure at a corner.

The singularity analysis of the material corners appearing at the samples corresponding to pull off and shear torsion tests, has shown that all the bimaterial corners have weak stress singularity values, and only the concrete corner at the bottom of the trepanation is

relevant, but has not affected significantly the failure initiation of the samples. A generalized failure occurring at any of these corners could have induced premature failures at these points and could have hidden the real strength values associated with the test.

With reference to lap peeling and lap shear tests, they can be properly described by a simple analytical beam model, which can be used to adjust some properties (including toughness) in order to develop more sophisticated models involving damage, using FEM. FE models have shown that the actual conditions in both lap peeling and lap shear tests corresponded to mixed mode of fracture, the lap peeling tests being near a mode I ($G_{CP} \approx G_{IC}$) and the lap shear tests being really close to mode II ($G_{CS} \approx G_{IIC}$), all this for the combinations of material systems considered in this research.

Failures were cohesive in all the tests performed. Thus, concrete strength and toughness would define the properties of the joint studied in this work.

For the carried out lap peeling tests, failure was toughness controlled, thus the fracture critical value G_{CP} of the concrete can be obtained either measuring the ratio of energy released for unit of new crack surface created, or adjusting the predictions of the corresponding beam model, avoiding in this way the necessity to measure the crack length during the test (a task that usually is not easy to carry out in a reliable manner).

However, failure is strength controlled for the performed lap shear tests. This happened because there was not sufficient bonded length for the crack to progress according to the classical Fracture Mechanic concepts, and consequently G_{CS} properties could not be measured. Thus, experimental results obtained from these tests could only be used to ensure a minimum value of fracture toughness which can be of enormous help when designing a bonded joint under dominant shear.

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Mechanical Joining of Stacks

Antonio J Gamez, Severo R Fernandez-Vidal, Alvaro Gomez-Parra, Pedro F Mayuet, and Ana P Valerga, University of Cadiz, Cadiz, Spain

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Glossary

Carbon Fiber Reinforced Plastics (CFRP) Composite material made of a reinforcement of carbon fiber and a matrix which could be made of a thermoplastic or thermoset polymer.

Ceramic Matrix Composite Materials (CMC) Composite material made of a ceramic matrix and reinforcements which are usually fibers of metallic, ceramic or organic origin, such as carbon.

Fiber Metal Laminate (FML) Layers of metal alloys and polymers reinforced by fibers through adhesive bondings.

Hybrid structure Assembly of materials of different nature, composites and/or alloys, in the form of sheets or plates, joined mechanically or by an adhesive.

Laminate Structure consisting of the assembly of layers of the same or different materials, generally bonded by adhesives.

Metal Matrix Composite Materials (MMC) Composite material made of a metal matrix and reinforcements which could be ceramic or organic compound.

Plastic Matrix Composite Materials (PMC) Composite material made of a polymer matrix (thermoset or thermoplastic) and reinforcements which could be fibers of different kinds.

Rivet Metal piece, also known as bolt or fastener, which serves to join different parts.

Stack Structure consisting of the assembly of layers of the same or different materials, generally mechanically bonded.

Introduction

Nowadays, industry demands materials with specific properties that can be hard to find in nature (Pereira and Romero, 2017; Ramulu *et al.*, 2001). Therefore, research is directed towards the design of new materials and the physical and chemical treatment of existing materials or their assembly to overcome their limitations and improve their behavior.

The combination of layers of the same or different materials with diverse configurations of composition, shape and thickness, seek the combination of advantages of the individual layers, and the elimination or reduction of some of their disadvantages (Xu *et al.*, 2016). In general, the assembly of hybrid materials, if compared to monolithic structures, provides better material properties, since the desired aspects of each constituent material are used and its weak points are avoided. Normally, the main advantage of this stacking is the combination of tensile strength, with the improvement of some specific characteristic, without a significant increase of the piece weight (Zitouni *et al.*, 2016). Therefore, a key application example is aircraft structures under high thermomechanical stresses, where mechanically bonded metallic materials and fiber-reinforced composites are used.

This article is focused on a particular combination of layers of materials called stacks. A stack is composed of different sheets or layers of materials, being identical or not, usually assembled on a particular direction in space, which results in a fairly compact piece depending on the type of fixation that holds them together. There are several terms used to refer to the combination of materials. However, the terminology is not consistent throughout the literature, and names are interchangeable in many cases (Sun *et al.*, 2014). Consequently, for ease of use, the following definitions will be used along this article:

- Hybrid structures refer to overlaps of materials of different nature, composites and/or alloys, in the form of sheets or plates, joined mechanically or by an adhesive. In the literature, this term usually refers to any structure formed by a combination of different materials, not necessarily stacked (reinforced with particles, nanotubes, etc.) (Xia *et al.*, 2017).
- Stacks refer to a particular way of disposing layers of materials. It can be used to refer to a set of sheets that may or may not be of different materials (Zhu *et al.*, 2018). Despite the breadth of this definition, it is usually more tied to arrangements that have mechanical instead of adhesive bonding. Even in some cases, the union is only given to take advantage of industrial (increase in production).[AJG5]
- Laminates consist of a series of overlapping sheets and can refer to layouts with sheets of the same material. This term is also used to refer to compounds that have been formed by superimposing layers of unidirectional fibers with the same or different orientation. In contrast to what has been said for stacks, laminates usually identify materials whose components or sheets have a non-mechanical bond, generally adhesive or a common matrix.
- Fiber Metal Laminates (FML) consist of thin layers of high strength metals and unidirectional fiber composites embedded in an adhesive system. The above concept can be extended to multidirectional fibers too. A polymeric matrix must be used as a fiber binder and, in some cases, also as adhesive bond between the alloy and the fiber composite. In short, FMLs are multi-component materials, usually in superimposed sheets, using metals, fibers, resin matrices and adhesives (Thirukumaran *et al.*, 2018). Unlike the previously defined terms, the FML is conceived as a whole, rather than as the integration of several differentiable elements due to the very process of manufacture. Also, layer thicknesses are often lower than those grouped under the categories of stacks and hybrid structures (Ansari *et al.*, 2018).

As for their assembly, whether stacking dissimilar materials or not, layers are normally joined by means of a mechanical fastening technique, which is the main method currently used for the assembly of structural components (Camanho and Matthews, 1997). This method has many advantages such as good reliability, interchangeability due to their easy disassembly, and convenient inspectability (Özaslan *et al.*, 2018; Kolesnikov *et al.*, 2008).

However, the key problem that arises through the use of mechanical fasteners is the high concentration of stress around the holes, which are also much more severe in composite laminates when compared to metal plates under the same loading conditions (Chowdhury *et al.*, 2015). This is mainly due to the properties of materials, as metals are ductile and can yield (Hart-Smith, 2001). In addition, the coupling between materials with a very different electrochemical potential causes an electrical connection, which creates conditions of high risk of galvanic corrosion (Serdechnova *et al.*, 2014). Therefore, the combination of a predominant mechanical fixation with adhesion is increasingly common in order to protect against these effects, especially stress concentrations that lead to premature or catastrophic failure when the elements are subjected to fatigue loading (Kelly, 2006). For example, in the aeronautical industry, this has allowed the certification of some of the aircraft structures.

Mechanical fixation is the one that guarantees the most hermetic union of multiphase materials, an essential requirement for structural parts. For this reason, the different phases of the materials that make up a stack are normally processed with a thorough quality control that preserves the integrity of the whole piece and of each part separately. The process for mechanical joining is the drilling, which can be done in a single step (One Shot Drilling, OSD), or at each stage of the stack separately, followed then by the assembly phase. The OSD accelerates the production process, minimizes positioning errors and favors tight tolerances in real production (Alonso-Pinillos *et al.*, 2019).

In short, section "Classification of Stacks" starts with the classification of hybrid structures according to the material. In section "Mechanical Joints", the different types of joining processes, mechanical, chemical and thermal are depicted, together with their advantages and drawbacks. In section "Mechanical Joining Processes of Structural Materials", mechanical joining processes are explained and, finally, some of the main sectors where this type of structures are found will be exposed in section "Industrial Applications of Hybrid Structures".

Classification of Stacks

In general, the four large families of materials: metallic alloys, plastics, composites and ceramics, are susceptible to being joined by stacking. In this section, the more widespread combinations of materials and joining processes used for industrial purposes are discussed.

Composites are formed by two phases: a continuous phase, called matrix, and a dispersed phase called reinforcement. Generally speaking, metals are more dense than ceramics and polymers. They are also rigid, resistant and ductile, which justifies their use as structural elements. Polymers, on the other hand, exhibit low density and high flexibility, but low strength. Ceramic materials are very rigid and as resistant as metals but have a very high fragility. Nevertheless, they are very good thermal insulators. For this reason, the action of these materials as stacks focuses on applications in which there is a large thermal gradient as, for example, space rocket coatings.

According to the type of matrix, a common classification for composites is the following: Metal Matrix Composite Materials (MMC), Plastic Matrix Composite Materials (PMC) and Ceramic Matrix Composite Materials (CMC) (Rajak *et al.*, 2019). In the case of PMC, its matrix can be thermoplastic or thermoset. Thermosets are more popular in use than thermoplastics due to their higher strength and resistance to high temperatures (Rajak *et al.*, 2019).

Stacks combine the same or different materials produced in possible diverse manufacturing process and exhibiting, in general, better mechanical properties due to the combination of the different physicochemical properties of each individual component. The design of hybrid materials is directed towards the increasing of mechanical properties and functionality (Muflikhun *et al.*, 2019). Stacks build of composite/metal show a high bending rigidity by the combination of materials as CFRP/titanium, CFRP/aluminum, for example (Pramanik *et al.*, 2017).

As stacks are composed by different layers, they must be joined. There are many definitions of joining, being the most suitable for our purposes the proposed by Martinsen *et al.* (2015):

- Mechanical – A joint formed through a mechanical mechanism
- Chemical – A bond formed through chemical reaction
- Thermal – A bond formed through application of thermal energy

Mechanical Joints

Bolted, fastener or riveting joints

Bolted joints are used to connect materials in order to form mechanical structures. In comparison to adhesive bonding and welding methods, bolted joints have been widely used due to their high load-carrying capacity, ease of assembly, and relatively low cost. Also, they facilitate the part replacement, repair, and maintenance and can be movable or adjustable.

The main disadvantage of bolted joints is the formation of high stress concentration zones at the locations of bolt holes, which might lead to a premature failure of the joint due to net-section, shear-out, or bearing failures, or their combinations

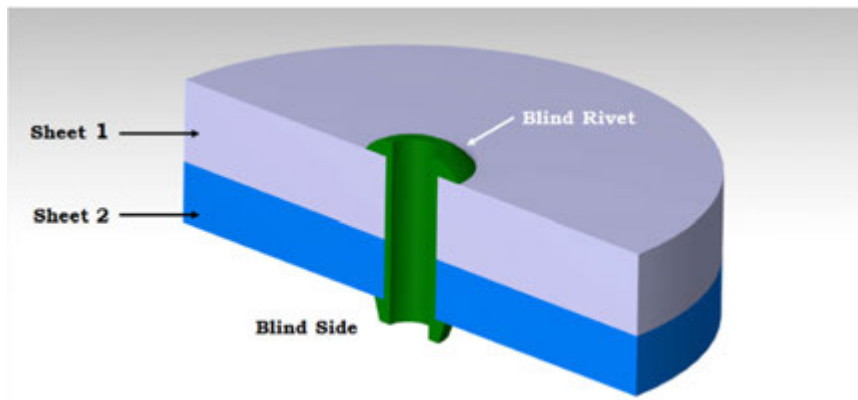


Fig. 1 Example of blind rivet.

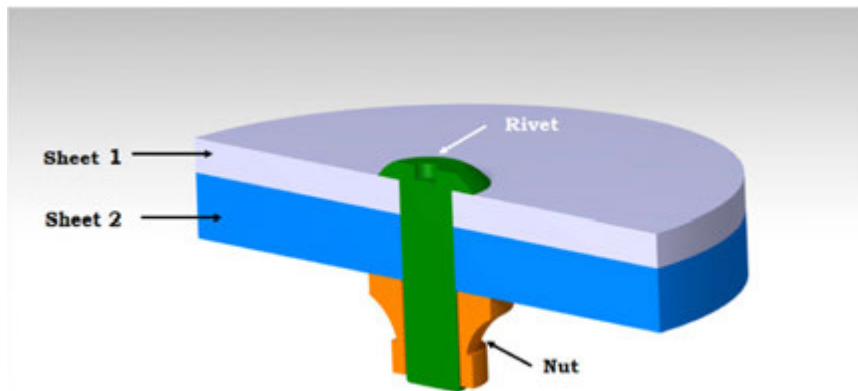


Fig. 2 Example of two – piece riveting.

(Kradinov *et al.*, 2007). For this reason, bolted joints can be considered the weakest location in the structural element (El-Sisi *et al.*, 2018). Joints can be formed by plastic deformation of the elements, but they are usually added to the structure by drilling of the layers.

Blind riveting

Blind rivets are used in areas where there is no access to the rear (blind side) of the joint. As main features they are hollow and have no thread. This geometric aspect is fundamental to allow their characteristic installation process. The hollow rivet body is crossed by a cylindrical rod, the stem, that has a head of greater diameter than the hollow body of the rivet. When the stem is pulled, the rivet body is deformed, generating the riveting action.

This type of riveting is one of the most used in the aeronautical industry and is present in the union of various types of stacks such as: metal-metal, CFRP-CFRP, metal-CFRP. An example of stack by blind rivet is shown in Fig. 1.

Two-piece riveting

Two-piece rivets are composed of a bolt, a nut and a washer to perform their clamping purpose. This type of joint has the advantage over the previous one of being removable. In this case, access to both sides of the joint is necessary. An example of stack joined by two – piece rivet is shown in Fig. 2.

Self-piercing riveting

This is a cold mechanical process used to join two or more sheets of materials by driving a rivet piercing. In this process, the rivet penetrates from the top to the bottom creating an interlock between the sheet of materials, see Fig. 3. This manufacturing process is used to bond different layers of materials such as aluminum alloys, titanium alloys, steel as well as composite materials. This process has many advantages because it does not require prior drilling, is easy to automate, does not generate chips or burrs, requires a low energy consumption, and generates different material joints with characteristics such as high strength and high fatigue performance (Di Franco *et al.*, 2013; Falk and Jäckel, 2019; Haque, 2018; He *et al.*, 2016; Li *et al.*, 2019; Pickin *et al.*, 2007;

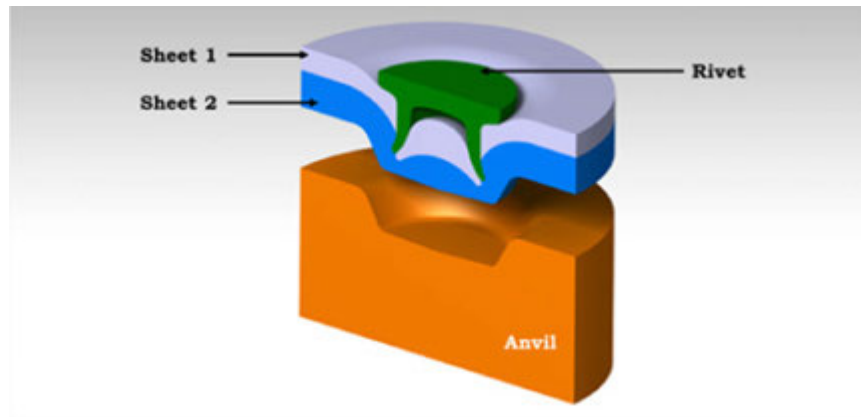


Fig. 3 Joint by self-piercing riveting.

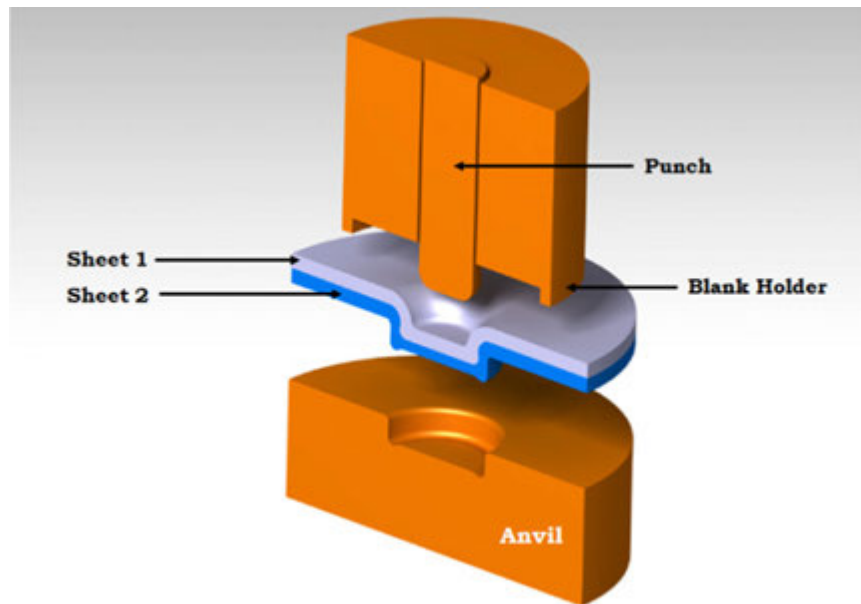


Fig. 4 Clinched sheets.

Zhang *et al.*, 2016). Self-piercing riveting requires access to both sides of the joint and the button created by the joint may not be esthetically acceptable (Haque, 2018).

Clinching

Clinching is a cold forming process suitable for joining dissimilar materials. With it, two or more sheets of different materials are locally deformed to create the joint, see Fig. 4. For this reason, there are no additional parts such as bolts or rivets needed in the process. A punch, a blank holder and an anvil are required as tools for clinching (Chen *et al.*, 2019b; Gerstmann and Awiszus, 2019). This process is widely used for the union of different metals in the automotive sector (Meschut *et al.*, 2014). However, there are studies showing that it can also be utilized for the union of CFRP to metals. As disadvantages, clinched joints have lower static strength than riveted joints (Chen *et al.*, 2019b).

Flow drill screws

This joining technique combines flow drilling and thread forming in a single procedure. In the process, a screw with a special tip drills stacked materials, first generating a plastic deformation process by extrusion. After this, a stage of thread forming takes place. Finally, a full thread engagement and tightening occurs when materials cool down.

Flow drill screw was developed as an innovative single-sided fastening solution for light weight thin sheet joints. As advantages, only one side access is required, pre-drilling of the materials is not necessary, can be used with adhesives in the interlayer of the

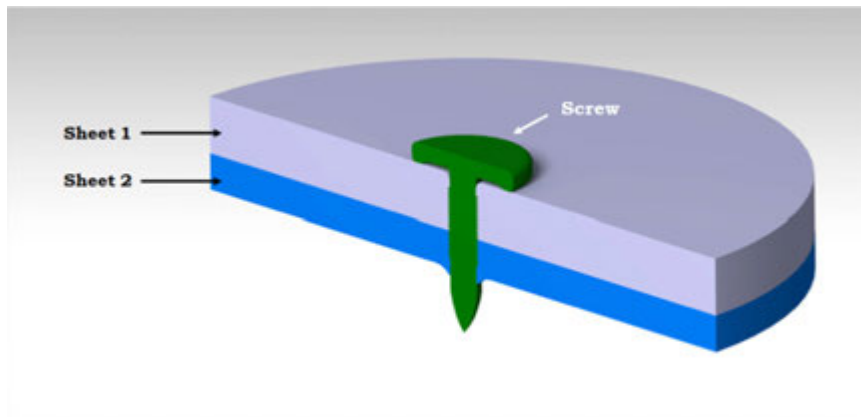


Fig. 5 Sheets of materials joined by flow drill screw.

materials to be joined, consumes low energy, and allows easy automation. This joining process has limitations when used with thin gauge materials because, to avoid skidding, the perpendicularity of the screw is needed (Graf *et al.*, 2018; Kumar and Jesudoss-Hynes, 2019; Martinsen *et al.*, 2015; Sønstabø *et al.*, 2018). In Fig. 5 a joint by flow drill screw is shown.

Chemical Joints

Adhesive bonding

Adhesive bonding is the process of binding two components using a suitable material (i.e., an adhesive). Adhesive joints act better than riveted joints because stresses do not concentrate around a given area as in the case of riveted joints. Moreover, adhesives can prevent corrosion acting as sealants, reduce mass in assemblies and reduce vibrations (Chowdhury *et al.*, 2015; Martinsen *et al.*, 2015).

Adhesives can be of natural or synthetic origin. Natural adhesives, also called bioadhesives, can be of vegetal or animal origin. Synthetic adhesives can be thermoplastics (cellulose acetate, polyvinyl acetate, phenoxy, etc.), thermosets (cyanoacrylates, epoxy, acrylic, etc.), or silicones (polyisobutylene silicone neoprene, etc.) (Ebnesajjad and Landrock, 2015).

Among the disadvantages of adhesive joints are: poor adhesions in some materials, low resistance, degradation, high inspection cost, very expensive or impossible repairs, extra time for polymerization or need of pre-processing of the surfaces. Roughly speaking, three stages can be identified (Oldewurtel, 2019):

- Mechanical treatment: They produce effects such as the removal of surface contaminants, creation of an active surface and increased contact area with the adhesive
- Degreasing: Procedure for surface decontamination by various solvents to obtain a correct degreasing, in order to obtain a perfect contact between the substrates to be bonded and the adhesive.
- Edge deburring: A smooth edge of elements to be bonded is one essential of good adhesion. Sharp edges are unfavorable for the roughness in the edge area may furthermore inhibit adhesion by preventing intimate contact between adherends.

Diffusion bonding

Diffusion bonding (DB) processes take place when the welded parts are in close contact under a controlled pressure and then heated to a defined temperature and for a specific time. These conditions allow atomic diffusion between the two welded parts, creating a high strength joint (Pramanik *et al.*, 2017). This joining process is extensively used for the joints of metals and in the joint of aluminum alloys an CFRP using interlayers of wires or foils of titanium and glass fibers (Pramanik *et al.*, 2017).

Thermal Joints

In this case, as in the fields of mechanical and chemical joints, only welded joints that allow the assembly of hybrid structures through stacking will be discussed.

Resistance spot welding

Resistance spot welding (RSW) is a high efficiency, low cost, robustness, flexibility, and widespread manufacturing process used in the join of similar metal in automated manufacturing (Chen *et al.*, 2019a).

This manufacturing process is very simple and is based on the use of two copper alloy electrodes that hold the sheets of metal together. After this, a welding current passes through the electrodes and metal sheets, melting and clamping them, see Fig. 6. This process is used to join different metals as aluminum alloys, copper alloys, magnesium alloys, steels or titanium alloys. Nevertheless, there are problems in the welding of aluminum alloys due to the generation of an oxide surface layer, and problems with

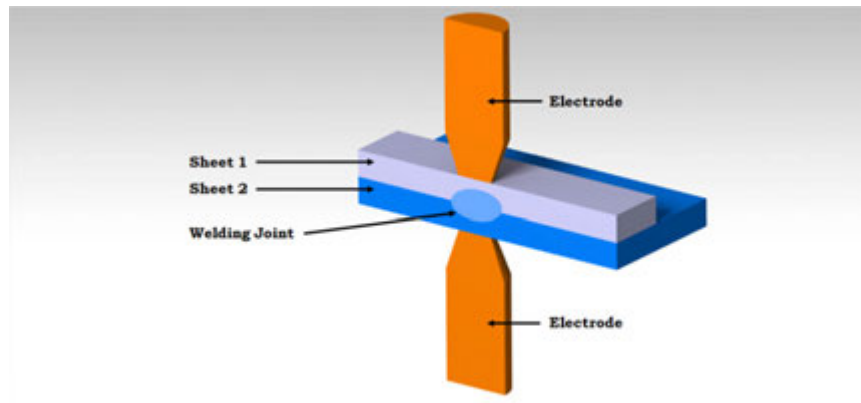


Fig. 6 Sheets joined by resistance spot welding.

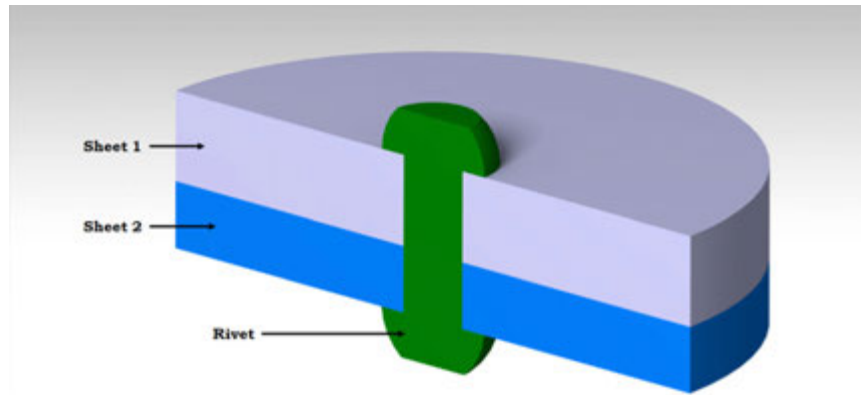


Fig. 7 Sheet metals joined by hot upsetting.

the different resistance in dissimilar materials (Al-Steel, for example) (Arghavani *et al.*, 2016; Chen *et al.*, 2017; Martinsen *et al.*, 2015).

Hot upset rivet

Hot upset riveting is a hot forming process that uses precision heat and pressure to form hardened workpieces. The installation of bolts in the assembly of materials is done by plastic deformation of one of the ends to create a second head of the bolt. This process achieves robust joints and, depending on the part requirements, the result can be a fixed or movable joint assembly.

This type of riveting is generally used in the joint of metals stacks including wolfram, titanium, stainless steel, carbide, molybdenum, hardened steels and other hardened metals. The disadvantage is the inspection of the joint and the difficulty of disassembly (Collette *et al.*, 2014), see Fig. 7.

Friction stir welding

The basic principle of friction stir welding (FSW) is fairly simple. In this process, a non-consumable rotating tool with a larger diameter shoulder and a pin, made of a material that is stronger than the workpiece, plunges into the workpiece to a pre-programmed depth. In Fig. 8, an example of join by friction stir welding is shown. The tool plunges between two sheets materials and due to frictional heat localized plastic deformation occurs in the joint section. Finally, the sheets are joined by a solid state welding process (Gite *et al.*, 2019; Padhy *et al.*, 2018; Patel *et al.*, 2019). This process by itself is not suitable for making joints of stacked materials. For this reason, several variations have been developed and are discussed below.

Friction self-riveting welding

This joining process, friction self-riveting welding (FSRW), is a variant of friction stir welding. In this case, a rotary tool without stir pin presses a metallic surface, generating frictional heat, Fig. 9. The sheet of material below must have prefabricated holes. The action of the tool melts the upper sheet introducing material from it into the holes placed in the sheet below. This process is indicated to join metals – metals or metals with polymer matrix composites and plastics (Huang *et al.*, 2019; Meng *et al.*, 2019).

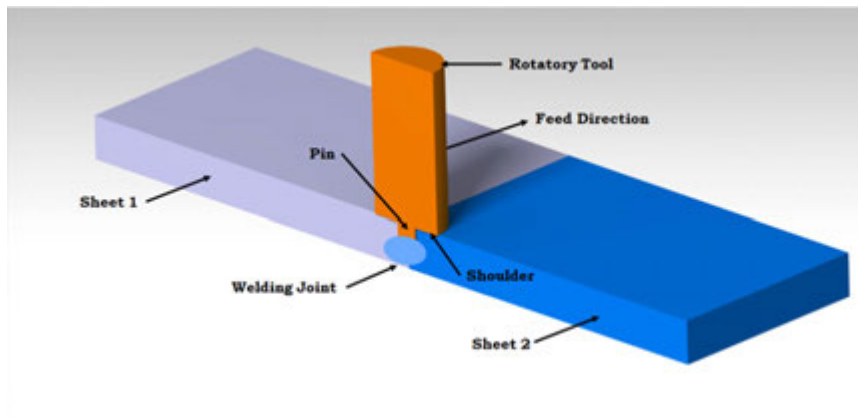


Fig. 8 Sheets joined by friction stir welding.

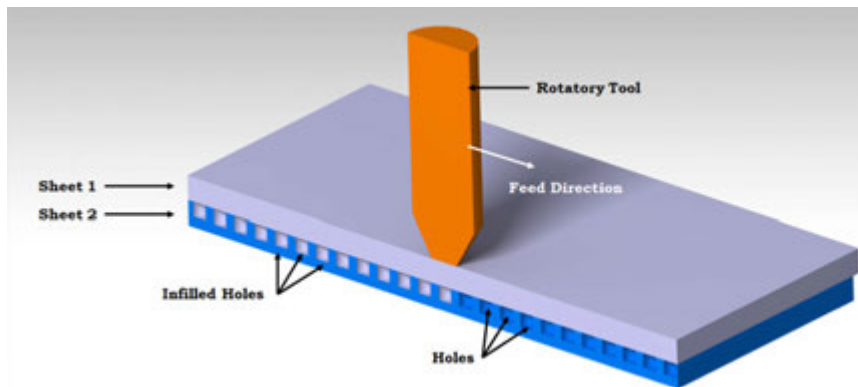


Fig. 9 Joint by friction self-riveting welding.

Friction stir spot welding

The friction stir spot welding (FSSW) process consists of three stages: plunging, stirring and retracting. The technique uses a three-piece non-consumable tool, comprising a pin, a sleeve and a clamping ring. The pin and the sleeve can move and rotate independently of each other while the clamping ring is the stationary part (Li *et al.*, 2019; Yang *et al.*, 2014). This joining method is able to joint metal-metal and metal-CFRP (Padhy *et al.*, 2018; Pramanik *et al.*, 2017), see Fig. 10.

Friction lap joining

The friction lap joining (FLJ), Fig. 11, process is recommended to join metal/metal and metal/polymer hybrid structures. The process is very similar to the friction stir welding, being the only difference that FLJ does not use a stir pin. The welding tool of FLW only has a rotating shoulder without the rotating pin (Huang *et al.*, 2018; Mishra and Sidhar, 2017; Wu *et al.*, 2018; Zou *et al.*, 2017). In FLJ, friction heat between the first layer of material and tool is transferred into the second layer, resulting in the melting of the interface, and then the joining different layers is achieved under the pressure of the tool and clamping tooling (Wu *et al.*, 2018).

Friction bit joining

Friction bit joining (FBJ) was developed to join dissimilar metal as Al alloys and steels. A rotary tool presses over a bit that is placed over two overlapped metal sheets. The bit is displaced from the upper sheet to the bottom. The rotating bit cuts through the alloy sheet and then engage the steel bottom sheet. Frictional heat and applied pressure soften the materials. Finally, the rotating tool stops, leaving the bit metallurgically bonded to the bottom material and mechanically fastened to the top material (Haghshenas and Gerlich, 2018; Martinsen *et al.*, 2015; Mehta, 2019), Fig. 12. A variant of this process is the Friction Self-Piercing Riveting (FSPR), in which a rivet like the one shown in Fig. 3 is generally used.

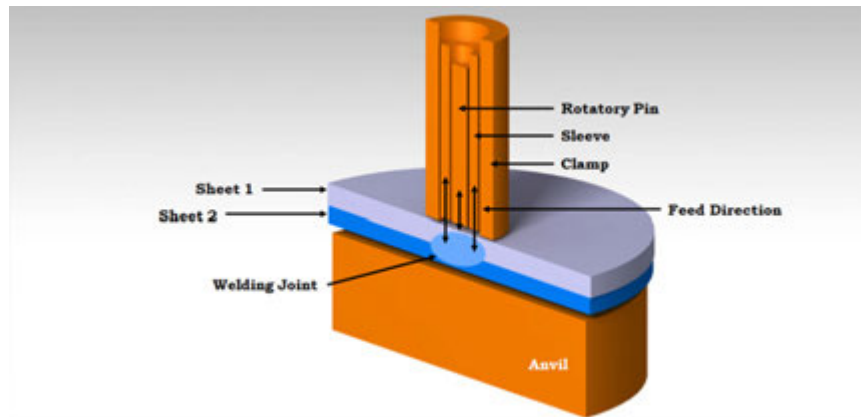


Fig. 10 Joining by friction stir spot welding.

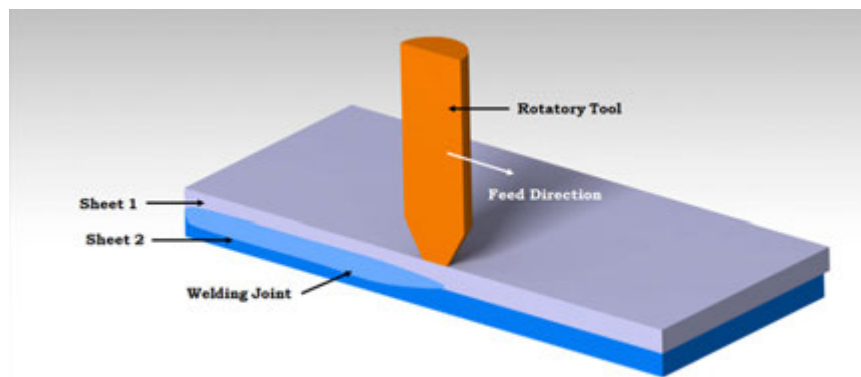


Fig. 11 Joining by friction stir lap welding.

Ultrasonic spot welding

Ultrasonic spot welding is a solid-state joining technology that uses a ultrasonic high frequency vibration energy to generate friction between the two faces of a sheet, thus achieving the bond by plastic deformation and softening of the material contact points (Li *et al.*, 2019; Ni and Ye, 2018). The temperature in the ultrasonic welding process does not exceed the melting point of the metal workpiece, eliminating undesirable compounds, phases and metallurgical defects (Martinsen *et al.*, 2015). Ultrasonic spot welding is pollutant-free, highly efficient, with a short weld time (typically under half second), and insensitive to material conductivity and heterogeneity (Ni and Ye, 2018). This joining process is capable to joint very different materials (Lionetto *et al.*, 2018; Ni and Ye, 2018; Wagner *et al.*, 2013; Wang *et al.*, 2020), see Fig. 13.

Mechanical Joining Processes of Structural Materials

The process of installing a mechanical joint can basically be summarized in the following sequence of operations (Campbell, 2006a,b):

- (1) Positioning and fixing of the parts to be joined in the assembly tools. The elements are placed in their final position to then be fixed by means of guide holes in which the temporary fasteners are inserted.
- (2) Drilling operations for the mechanical joints inclusion.
- (3) Hole quality control. Surface quality and dimensional and geometric tolerance of the drill are checked to verify they are adequate for the type of mechanical joint selected. Checking operations are normally carried out visually and with gauges.
- (4) Disassembly of the pieces. At the end of the drilling process, all the fixing elements are removed and the tool components are disassembled.
- (5) Cleaning and repairing of defects. Absence of dust or shavings in the interface and absence of burrs in the holes in metallic materials, or delamination in the fiber of composite materials are checked. If this is not the case, cleaning, deburring and repairing of delamination are carried out.

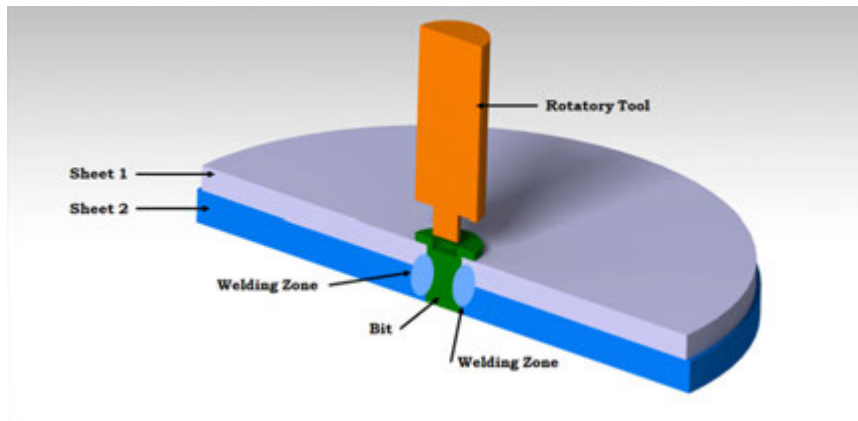


Fig. 12 Friction bit joined sheets.

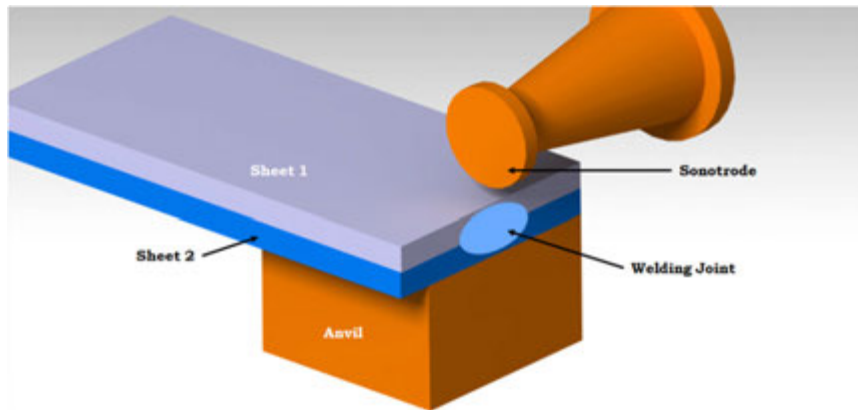


Fig. 13 Joining by ultrasonic spot welding.

- (6) Sealant application. After the cleaning and repairing operations, sealant is applied to the shells to ensure watertightness and increase resistance to corrosion.
- (7) Repositioning of parts on assembly tools. The parts are put back into the tooling in their final position.
- (8) Final clamping operation. The pieces are fixed with fasteners through the holes.
- (9) Quality control of fasteners. Correct positioning of the fasteners is verified.

However, this installation process can be very time-consuming, given the large number of fasteners a structure has, such as is the typical case of an airplane. Thus, alternatives have emerged in the industry, aimed at optimizing this assembly process by mechanical joints (Campbell, 2011).

The main optimization proposal would be to automate the assembly process. However, this approach faces difficulties, among them the variability in geometry and dimensions of the parts to be joined and the tight geometric and dimensional requirements to be met in this assembly. Therefore, in case of materialization, a high investment in the necessary equipment would be required (Grote and Antonsson, 2009; Stalley, 2002).

An alternative worth mentioning is the assembly technique known as One Way Assembly (OWA). This technique aims to ensure quality in the different operations involved in the process without having to separate the components to verify the results. This would imply the elimination of steps 3, 4, 5 and 7 in the previous sequence, Fig. 14. The elimination of these steps does not preclude action in case the quality requirements are not met. That means, actions such as the cleaning of chips trapped in the interface, repair of delaminations or removal of burrs will be carried out if necessary. This requires the establishment of a system of verification or constant inspection during the process, reaching to require a certain degree of automation. In addition, the use of cutting fluids must be eliminated in order to avoid the separation of components. This makes the OWA technique an environmentally friendly assembly methodology (Stalley, 2002).



Fig. 14 Stages of the OWA Mechanical Joining Process.

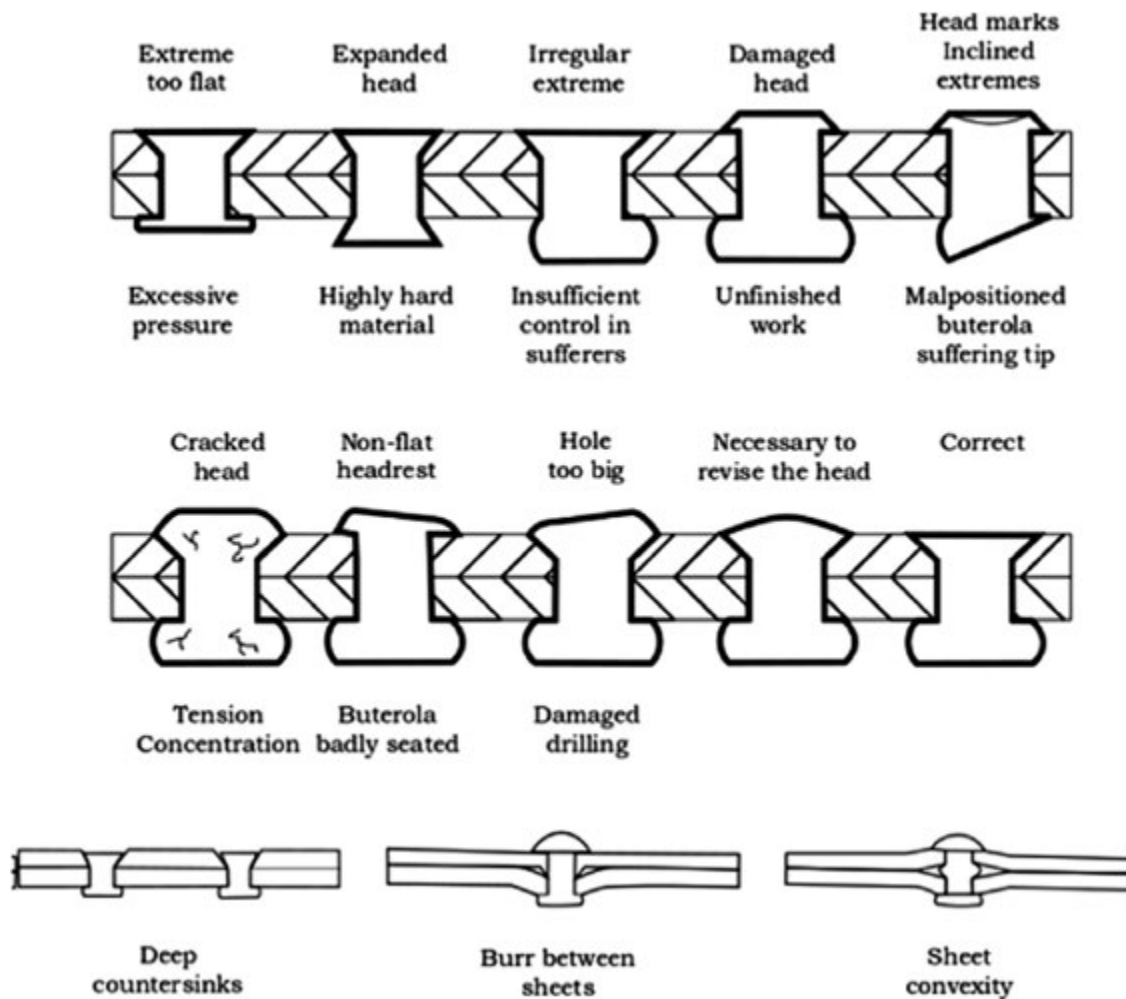


Fig. 15 Defectology in mechanical joints.

Defectology

There are other aspects to consider during the assembly process which, if not taken into account, would negatively affect the joint, as the existence of chips or burrs in the machined drill, or simply, the inaccurate situation of the elements during the installation process. Consequently, in aeronautics and other sectors where riveted joints are the main assembly method, the installation process is considerably more complex, and many different types of defects appear, as shown in [Fig. 15](#).

Defectology due to the nature of the material

In the case of composites, the most critical defect generated by drilling is delamination. Delamination occurs mainly at the entrance and at the exit of the drill, when significant axial forces are generated during machining, resulting in the separation of the material faces ([Campbell, 2006a,b](#); [Davim, 2018](#)), as a result of the fact that the mechanical properties of the matrix are weaker than those of the reinforcement, [Fig. 16](#). Although the mechanisms of formation of delaminations are similar at the entry and at the exit of the hole, the causes of their formation are totally different.



Fig. 16 Delamination in composite materials.

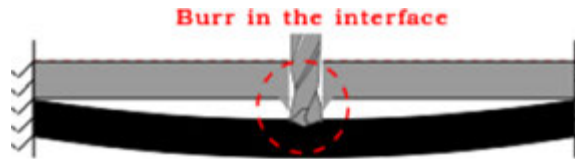


Fig. 17 Interfacial Burr Formation in Drilling of Stacks.

The delamination at the entrance begins when the tool contacts the first layer of material. It is produced by plastic deformation and twisting of the material as a consequence of the axial load and torque developed by the tool. In the first moments of machining, the edges do not have cutting capacity, causing the material dragging into the tool evacuation channels. This type of delamination is produced mainly by the forces generated during drilling, these being related to the material, the geometry of the tool and the cutting parameters.

The delamination at the exit is mainly caused by the cutting conditions imposed during machining. As a consequence, the consistency stresses of the final layer matrix are exceeded by the axial load developed by the tool. This defect is favored by the loss of cutting capacity of the edges due to wear, which increases the axial force provoked by the tool (Davim, 2018, 2015).

Chipping or fraying occurs, like delaminations, at the inlet and outlet of the drill as a result of the forces to which the material is subjected and the low flexural strength of the layers.

The geometric deviations are due to the anisotropy of the material. For each angular cutting position in relation to the orientation of the fiber, there is a different reinforcing direction, where the tool is subjected to compressive and tensile stresses (Persson *et al.*, 1997).

Thermal damage deteriorates the matrix due to a temperature increase by the friction generated by the interaction between the tool and the machined material during drilling (Hocheng and Tsao, 2005; Piquet *et al.*, 2000). This phenomenon occurs if the temperature exceeds the transition temperature of the matrix vitreous state (T_g). As the matrix is responsible for the cohesion of the fibers, if it suffers thermal deterioration, propagation of defects as delaminations, cracks or points with discontinuities of material occurs. The combination of low feed rates and high cutting rates in the drilling of composite materials produces carbonized resin damage on the surface of the hole surface (Persson *et al.*, 1997).

In the case of the drilling of metal alloys, burrs are the most important defects (Nichol, 2001; Costa *et al.*, 2009; Efstathiou *et al.*, 2017; Pilný *et al.*, 2012; Ferreira *et al.*, 2011). Burrs happen as a result of a series of fractures and plastic deformations produced on the uncut material at the periphery of the hole, and they are a consequence of the movements of the drill bit, Fig. 17. The final shape of the burr depends on multiple factors. Among these are the material, the geometry and the coating of the tool (Rivero *et al.*, 2006; Lauderbaugh, 2009; Melkote *et al.*, 2010), the feed rate between the cutting parameters (Rivero *et al.*, 2006; Giasin *et al.*, 2019), the high temperature generated in the cutting area (Rivero *et al.*, 2006), cutting conditions (Melkote *et al.*, 2010; Giasin *et al.*, 2019), part clamping (Lauderbaugh, 2009; Jie, 2013; Fernandez-Vidal *et al.*, 2018; Bañon *et al.*, 2019), tool wear (Cantero *et al.*, 2005), the part material (Nichol, 2001; Bañon *et al.*, 2019; Saunders and Mauch, 2001), and the ratio diameter-length of the hole (Nichol, 2001; Melkote *et al.*, 2010).

Defectology due to interference of the drilled material

During the drilling process, there is a continuous material removing and wear of the tool. These conditions can affect the cut and, accordingly, the geometric and surface quality of the hole. The final quality of the machined surface will consequently be affected by chip interference during evacuation to a greater extent when there is dissimilarity of materials (Fernandez-Vidal *et al.*, 2018; Bañon *et al.*, 2019).

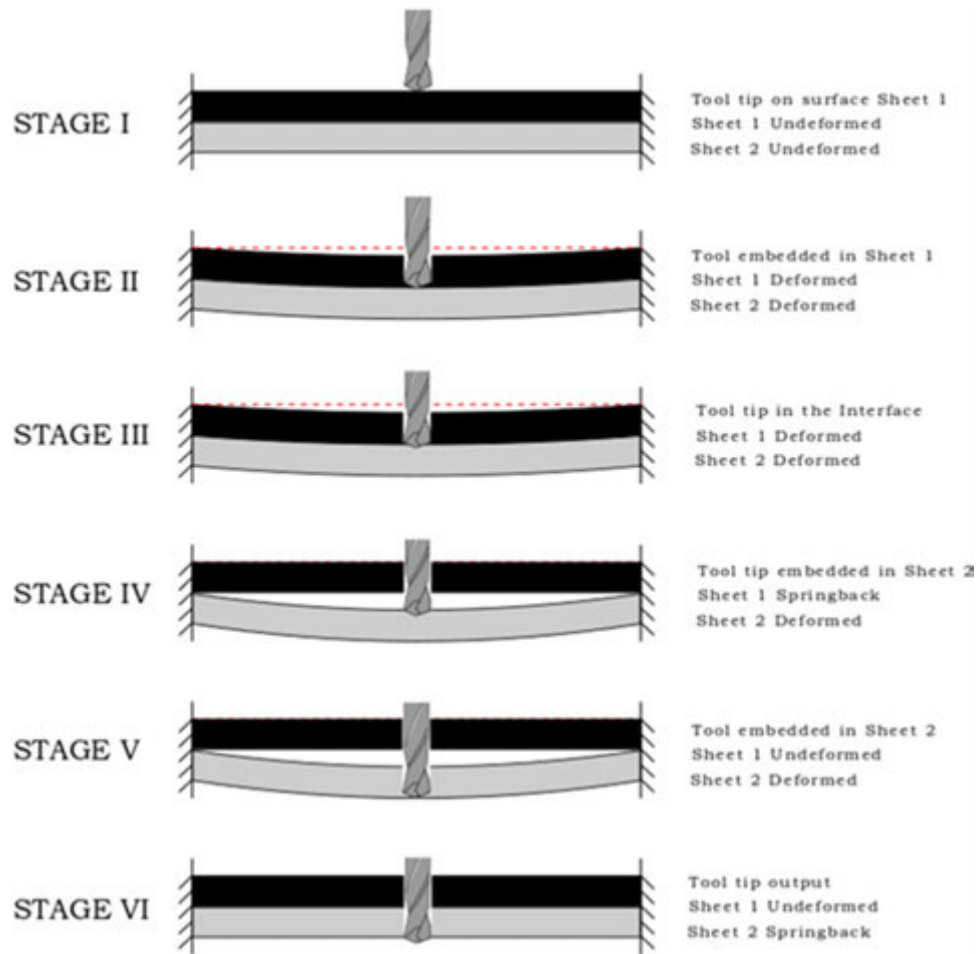


Fig. 18 Multi-stage drilling process.

Stacking defectology

In the stack drilling process, the thrust force developed by the tool can cause the stack to deform and even separate its sheets, [Fig. 18](#), affecting the quality of the machining ([Gao et al., 2015](#); [Wei et al., 2016](#); [Luo et al., 2019](#)). The deformation can be described by different stages of the stack drilling:

In stage I, the tip of the tool is located on the surface of the first plate.

In stage II, the first plate is drilled. Deflection occurs in both plates as a result of the axial load produced by the tool.

In stage III, the tool makes contact with the second plate. The thrust force of the first plate decreases, while that of the second plate increases. The deflection of each plate differs at the end of this stage.

In stage IV, both plates are being drilled. The deflection of the first plate is reduced to zero, while the deflection of the second plate continues to increase until the tip of the tool is not fully embedded. Separation between plates occurs.

In stage V, only the second plate is perforated. The deflection of this plate, as well as the spacing between both plates, reaches a maximum.

In stage VI, the second plate is drilled. Both deflection and plate spacing are reduced to zero.

Among the possible defects that can cause the deformation of the plates and their separation during the drilling operation, we can mention the loss of coaxiality between the holes of the different plates ([Gao et al., 2015](#)); the enhancement of the generation of delaminations or burrs ([Wei et al., 2016](#)); and the remains of material cut in the interlayer ([Luo et al., 2019](#)).

Defectology in the mechanical bonding process

Plastic deformation of the connecting elements via compression exposes the joints to a series of defects. The pressure exerted on the rivet can introduce high stresses on the components of the joint, a phenomenon even more pronounced when the assembled parts are not metallic. This can lead to cracking of the element or these assembled parts in the region close to the orifice. Also, excessive interference or expansion of the connecting element can lead to sufficient compression to cause defects in the element and in the material.

The pre-drilling operation to the installation of the joint element has a direct relation to the quality of the joint. Aspects such as the dimensional tolerance or the surface quality of the hole affect the joint greatly. In reference to the quality of the joint, the appearance of defectologies in the material, such as delaminations in composite materials or burrs in metals, lead to problems derived from excessive interference. However, it may be the other way around, i.e., the adjustment interference being insufficient as a result of a too large hole.

Industrial Applications of Hybrid Structures

The scope of application of hybrid structures is essentially linked to the diverse transportation industries. In the analysis of their objectives and technological challenges, research needs and difficulties for the years 2025–2030, reduction of weight and fuel consumption are among the main priorities, along with the decrease in costs of manufacturing to increase sustainability and environmental quality. In this sense, composite materials are more important for the aerospace, naval, space and automotive industry, where they join traditional materials in order to lighten weight and improve the final mechanical properties (Chavhan and Wankhade, 2019).

The implementation of hybrid unions has a significant relevance in responding to the main needs of the aforementioned sectors. For example, the (European Commission, 2011) has dedicated a budget of € 6.339 million for the 2014–2020 period to the Smart, Green and Integrated Transport work program in order to finance activities aimed at achieving efficient transport in terms of resource use and respectful with the environment, reaching better levels of mobility, less congestion and a greater degree of safety.

In addition, one of the most relevant programs of public-private in the aeronautical sector has been (Clean Sky 2, 2013). This program has as main objectives the reduction of emissions of gases, such as CO₂, produced by aircrafts, seeking to contribute to the intelligent and sustainable development of the sector in the space field.

Aerospace Sector

When it comes to selecting the material for structural components subjected to enormous efforts, the aerospace sector uses principally hybrid materials composed of CFRP and titanium (Kazemi *et al.*, 2019).

The increase in pieces of composite material in the latest generation of aeronautical projects has also led to an increase in the content of titanium parts. This is due to the greater compatibility between these two types of material in terms of galvanic corrosion. The joints of structural parts of Ti and composite material are currently made by riveting. This mechanical procedure, in addition to weakening the structure by drilling, also increases the weight, (Soutis, 2005). The main objective of this technology line is to glue titanium reinforcements in the CFRP structure of highly loaded areas. To achieve this objective it is necessary to study the following aspects: selection, development and optimization of the surface treatment of Ti, study and development of machining and drilling of hybrid materials, non-destructive inspection of hybrid materials, mechanical tests with different configurations and industrialization of the process (Kazemi *et al.*, 2019; Lachaud *et al.*, 2001; Ramulu *et al.*, 2001). The advantages of this technology are its weight saving and reduced production cost, as well as improved fatigue resistance and diminishing crack propagation.

In general, there has been a progressive increase in the replacement of metal parts by composite materials in military and civil aircraft. For example, composite materials represent more than 50% of the total weight in programs such as the Boeing 787, the Bombardier C series or the Airbus A350 XWB. In the latter, for example, the central wing box, the tail cone, the pressure bulkheads and the vertical and horizontal tails are manufactured with CFRP/Ti material joints, and this trend is likely to increase in the near future (Salguero *et al.*, 2015).

The combination of different materials to give rise to hybrid configurations in the design of high performance structural components is capable of satisfying multiple requirements in terms of specific stiffness and strength, as well as impact resistance and damage tolerance. For the joining of these different materials, mechanical bonding prevails, although it is true that the joining of these dissimilar materials can be carried out, both by means of the traditional riveting, and by using adhesive bonding techniques or the use of welding in the case of thermoplastic materials (Campilo *et al.*, 2018).

Although the need for implementation of joints between dissimilar materials is a fact today, its analysis and characterization are complex, since there is not enough knowledge and regulatory regulation to address this task. This makes the implementation of an experimental methodology for the mechanical characterization of adhesive joints between dissimilar materials one of the objectives of the aerospace industry by 2025, as well as the development of specific structural design tools with these joints. In this way, the trend marked by the sector is to eliminate laborious machining processes to allow an economical and safe assembly for the process.

Automotive Sector

In traditional car manufacturing, the use of metals and plastics is a matter of choice for various structural and non-structural applications. Currently, this paradigm is gradually changing with the introduction of polymeric-metallic hybrid structures, in which metals and polymers are integrated into a component (Hodkinson and Fenton, 2001).

In the automotive industry, stacked materials have been steadily increasing for several years because the trend in this industry is towards weight reduction in order to reduce energy consumption. Thermoplastic-metal hybrids are a viable alternative in which rigidity, impact resistance and functional integration are combined in a lightweight and cost-effective solution (Guo *et al.*, 2019). Metallic materials are used in areas where high rigidity and strength are required, while thermoplastic facilitates functional integration, thanks to the formation of complex geometries during the molding process. In this way, the combination of both materials optimizes the performance and functionality of the application in a specific way by balancing the contribution of metal and thermoplastic (Feraboli and Massini, 2004).

However, there are some obstacles that prevent the extensive use of polymer matrix composite materials in the transport sector, as their limited manufacturing cadence, high cost of materials and processes, and recycling/repair difficulties.

For this reason, the aim is to use metallic materials for areas requiring structural mechanical properties, and to combine this with the functional integration and geometric complexity that plastic allows (Haghshenas and Gerlich, 2018; Huang *et al.*, 2018). This becomes the standard, for example, in the supports of the front sections of vehicles, in which a combination of metal and plastic is used to form an upper crossbar that contributes to the rigidity of the car and is capable of supporting loads such as, for example, the closing of a latch. Another example is the use of metal-plastic hybrid structures through a composite material made of polypropylene with a load of long chain glass fiber, linked to a metallic reinforcement by means of adhesives to provide superior performance to other methods of forming hybrid systems (El-Sisi *et al.*, 2018).

It is expected that developments in the field of vehicle lightening will have the following consequences (Hodkinson and Fenton, 2001; Guo *et al.*, 2019):

- The weight of the vehicles will be reduced and, accordingly, the levels of consumption, emissions and the amount of raw material used to manufacture them will be reduced.
- Deterioration of public roads and noise will be reduced.
- The payload of vehicles will be increased, enabling commercial vehicles to increase their performance and efficiency per journey.
- The size of the parts used will be reduced and the habitability on board the vehicle will be improved.
- New complements and functionalities will be introduced without increasing the total weight of the vehicle.
- There will be a reduction in the wear of tires and brakes, lengthening the periods of change and, therefore, making car maintenance cheaper.
- They will allow a greater level of customization in the components allowing a greater degree of competitiveness based on the differentiation of product ranges.

Naval Sector

In the naval sector there is also an increase in the use of composite materials at a structural level, where a small percentage belongs to multi-layer hybrid materials made up of metal sheets, alternating with others made of composite material and structural adhesive, resulting in improved service performance (Gargano *et al.*, 2017). As a consequence, the necessary engine power of the ship is reduced and, accordingly, the pollution generated by it.

Hybrid fiber-metal laminated materials combine high impact resistance and durability, together with versatility in the production processes of metallic materials, with specific resistance and rigidity in the direction of the fibers, as well as good fatigue behavior (Li *et al.*, 2019). The multi-layer hybrid material used in structures is made up of layers of steel and composite material of vinyl ester matrix and glass fiber reinforcement. Through the combination of these materials, the precise positioning and orientation of each layer and the union of the different layers with each other by means of elastic structural adhesives, hybrid panels with superior performance than competing materials are achieved as a final product. The result is a lighter, stronger, tougher and safer material that can be custom designed to meet the specific requirements of each area of the structure (Mathijssen, 2016). The modular manufacturing process is capable of adapting to both flat and curved geometries and the assembly process ensures watertightness and correct load transmission from one panel to another, while improving tolerance to structural damage through mechanisms that stop the propagation of cracks.

The current challenges in naval research and technology focus on the development of platforms, conventional vessels, multi-hulls or modular surface effect vessels that will be more agile and environmentally more efficient in terms of lower emissions and fuel consumption, with a wide capacity for automated operation. This, combined with a reduced cost life cycle that includes intelligent diagnosis for lower maintenance cost, make hybrid materials able to meet these needs (Kharghani and Guedes-Soares, 2018).

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Mechanical Joining of Composites: Drilling Related Aspects

Juan M Vazquez-Martinez, Irene Del Sol, Jorge Salguero, and Moisés Batista, University of Cadiz, Cadiz, Spain
Carlos R Alcalá, Airbus Operations SL, Cadiz, Spain

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Introduction

Composite materials, especially Fiber Reinforced Polymers (FRP) had been widely used in the aerospace industry. Among them, Carbon Fiber Reinforced Plastic (CFRP) and Glass Fiber Reinforced Plastic (GFRP) are the most used ones, being up to a 50% of the Boeing B787 structure and up to a 53% of the Airbus A350 manufactured based on this materials (Gilpin, 2009).

These composite parts come from a nearly final shape but machining cannot be avoided especially for final size fit and assembly purposes (Feito *et al.*, 2016; Krishnamoorthy *et al.*, 2009). Regarding the assembly task, it is quantified as a 50% of the final part cost, and usually involve drilling operation. It can be performed using adhesive (cured and co-cured) alternatives, although traditional mechanical joints, such as screws, riveting, fasteners and bolted joints, are still the most commons for aeronautical purposes (Stocchi *et al.*, 2013; Lee *et al.*, 2017; Schürmann, 2007). They ensure an accurate assembly of the components and joining procedures after a highly control of their quality. The advantages of mechanical over adhesive joining or welding are (Akovali, 2001):

- (1) They met the standards of safety requirements, being more reliable in structural terms after inspection.
- (2) Better sustainability of the bonding process by the use of components based on environmentally non-toxic recyclable materials (aluminum, titanium, steel, etc.).
- (3) Ease assembly techniques with conventional mechanical tools without curation or drying time.
- (4) Increased process flexibility by the ability to repeated mount and dismount the joined parts, improving the verification, replacement and repair stage.
- (5) Lower sensitivity to environmental degradation. They have a better resistance to corrosive, thermal and environmental effects.
- (6) High strength of the assembled parts, especially for peel loading.

However, the interaction between three different elements may be subjected to adverse manufacturing conditions that may cause deviations on the final dimensions and geometry of the contact surfaces. Also, the use of metallic materials for the auxiliary assembly components and composite substrates as joining parts, may show lack of compatibility between assembled elements, increasing the corrosion effects, Table 1 (Campbell, 2006).

Other mechanical techniques, like clinching, also use a pilot hole and direct threat or self-piercing have been tested. However, the technology is not still mature and can increase part damage. It is more useful for metallic components (Nagel and Meschut, 2017) and its applications are right now mainly focused on automobile sector (Küçükoğlu and Karpat, 2017). Similarly, hybrid joining, combining adhesive and bolted joints, seems to be a promising future. They reduce the number of metallic components added to the structure and they significantly improve the performance of the adhesive joints (Pramanik *et al.*, 2017) but still, drilling operation will be need. This fact, remarks the importance of the drilling operation in terms of cost of the structure, especially for it assembly.

Bolts joints are fixed through a uniaxial hole drilled in the parts to be joint. Drilling is a high value operation due to the cost of the work piece in its final stages (Krishnaraj *et al.*, 2013). This operation can represent up to a 2% of the total cost of the structure and it should be considered that drills are performed on almost finished parts. In fact, poor quality of holes is considered the reason of up to a 60% of the part rejection (Hocheng, 2012) because this operation may be the cause to strength loss, and stress that have to be taken into account in the joining tolerances design.

Drilling of FRP has its own characteristics that make it a challenge operation. The diameter of the hole should be slightly smaller than the bolt and it may include countersunk shape or plain depending on the bolt geometry which will be seal with special sealant. A smaller hole will increase the friction between the surface of the part and increasing delamination, as well as increasing the surface roughness and that irregularities that may produce delamination and friction on the fasteners. Although, too big holes will ease the bearing failure producing load-displacement curves (Pramanik *et al.*, 2017). For this reason, the hole quality has also an impact on the joining and the anisotropy and the abrasive nature of the fibers makes the drilling process more complex (Faraz *et al.*, 2009). The surface quality of the machined edge, the cutting temperature or the coolant effectivity are mainly affected by the type of fiber reinforcement and its orientation (Sheikh-Ahmad, 2009).

The most frequent feature-related geometrical damages in FRPs are: delamination, fiber pull-out and uncut fibers, micro-cracking, inappropriate surface roughness and matrix burning (Geier *et al.*, 2019). Additionally, drilling of FRP part produces defects such as microcracking, fiber breakage debonding, matrix cratering, thermal damage and delamination among others.

Delamination could be considered as the most influence defect, playing an important role in the fracture or break of the joint. It takes place at the entrance and the exit of the hole, peeling it surface and affecting the residual stress and the behavior of the composite (Xiao *et al.*, 2017). For this reason, the measure of the quality of the hole is a key factor, and the detection of

Table 1 Compatibility between substrate and auxiliary joining component materials

<i>Substrates to be joined</i>	<i>Auxiliary join element material</i>	<i>Compatibility</i>
Aluminum - Aluminum	Anodized aluminum	Recommended
	Titanium	Acceptable
	A286 steel alloy Cadmium plated steel	Non-Recommended
Titanium-Titanium	Titanium	Recommended
	A286 steel alloy	Acceptable
	Steel alloy	Non-Recommended
Austenitic Stainless Steel-Nickel based alloys	Inconel 718	Acceptable
	Aluminum Aluminum coated fasteners	Non-Recommended
Titanium-Aluminum	Titanium	Recommended
	A286 steel alloy Inconel 718	Acceptable
	Aluminum Aluminum coated fasteners	Non-Recommended
Composite Carbon/epoxy	Titanium	Recommended
	Inconel 718 A286 steel alloy	Acceptable
	Aluminum Aluminum coated fasteners	Non-Recommended

delamination size could reduce joining problem during the service of the part. They can reduce stress concentration and inter-laminar crack propagation that enhance the probability of failure modes produced in the joint, some examples are given in [Fig. 1](#).

Apart from the hole quality, joining time can be increased depending on the characteristics of the operation, which rate is usually between 1.5 and 4 holes per minute ([Lambiase et al., 2017](#)). Even though typical drilling operation takes place in two steps, drilling and reaming. Most of the drilling advances focus on One-Shot-Drilling (OSD) and/or One-Way Assembly (OWA). These techniques also reduce the number of non-conformed parts due to hole location and reduce the number of temporary fasteners decreasing the operation time.

Hole Requirements

The manufacturing tolerance and accuracy of the results are highly related to the process selection as is shown in [Fig. 2](#). The process can be classified depending on the automation level of the drilling equipment, covering from manual devices to totally automated systems (CNC) ([Campbell, 2006](#)). Devices are chosen based on the nature of the assembled parts and a wide range of evaluation techniques to ensure its quality. The measures on the holes ensure the dimensional accuracy and minimize the macro and micro geometrical deviations regarding the nominal requirements.

The hole is evaluated in terms of geometry and integrity. The geometry of the hole covers a wide range of parameters with possible dimensional requirements whereas, hole integrity evaluates the surface finishing, overheating, contamination and damages (delamination, flaking or scoring). These requirements are based in experimental and theoretical studies, which provide the dimensional tolerances for the different manufactured parameters. The fits can be classified as a clearance fit or a transition fit.

When drilling composite layers, a maximum average roughness value of 1.6 μm must not be exceeded, and delamination have not allowed on the hole exit or entrance. Regarding to diameter specification, although aerospace structures usually takes as reference the H7 ($\pm 12 \mu\text{m}$) tolerance from ISO 286 standard, the drilled holes on composites may shows H8 ($\pm 18 \mu\text{m}$) or H9 ($\pm 30 \mu\text{m}$) ([Giasin et al., 2017](#); [SANDVIK COROMANT, 2010](#); [National Centre For Aerospace and Transportation Technologies, 2014](#); [Walczyk et al., 2000](#); [Caggiano et al., 2019](#)). Dimensional and geometrical deviations may be the main cause of undesirable

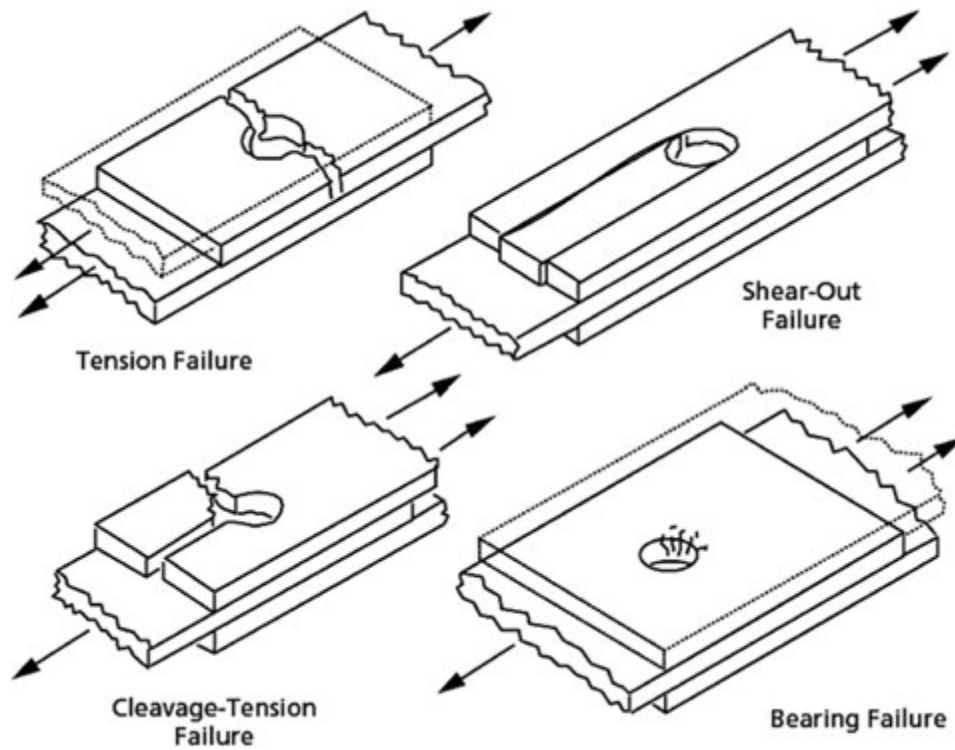


Fig. 1 Different modes of failure produced by hole defects. Reproduced from Campbell, F.C., 2006. Manufacturing Technology for Aerospace Structural Materials. Elsevier Science.

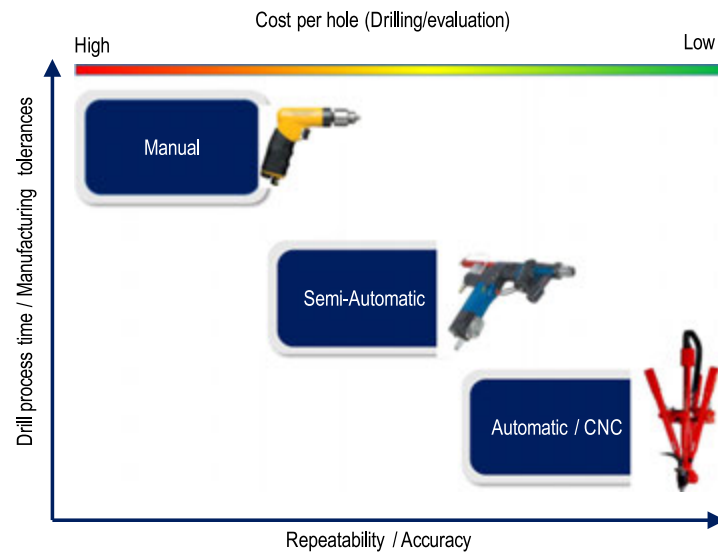


Fig. 2 Drilling process features depending on the equipment.

effects on the joining system, causing structural stresses and fatigue situations which can weaken the substrate material and decrease the safety of the assembled system.

The drilled holes are evaluated using metrological techniques based contact and non-contact measurements and additional visual inspections. On the one hand, the most used method for evaluate the dimensional size of the holes is to directly measure them using a three-contact micrometer. Nevertheless, more accuracy measurements can be achieved by non-contact devices, such as laser transducer or optical inspection.

On the other hand, surface finishing of the drills provides valuable information about the anisotropic properties of the composites and the drilling process. Incorrect roughness on the contact surface between the hole and the auxiliary assembly

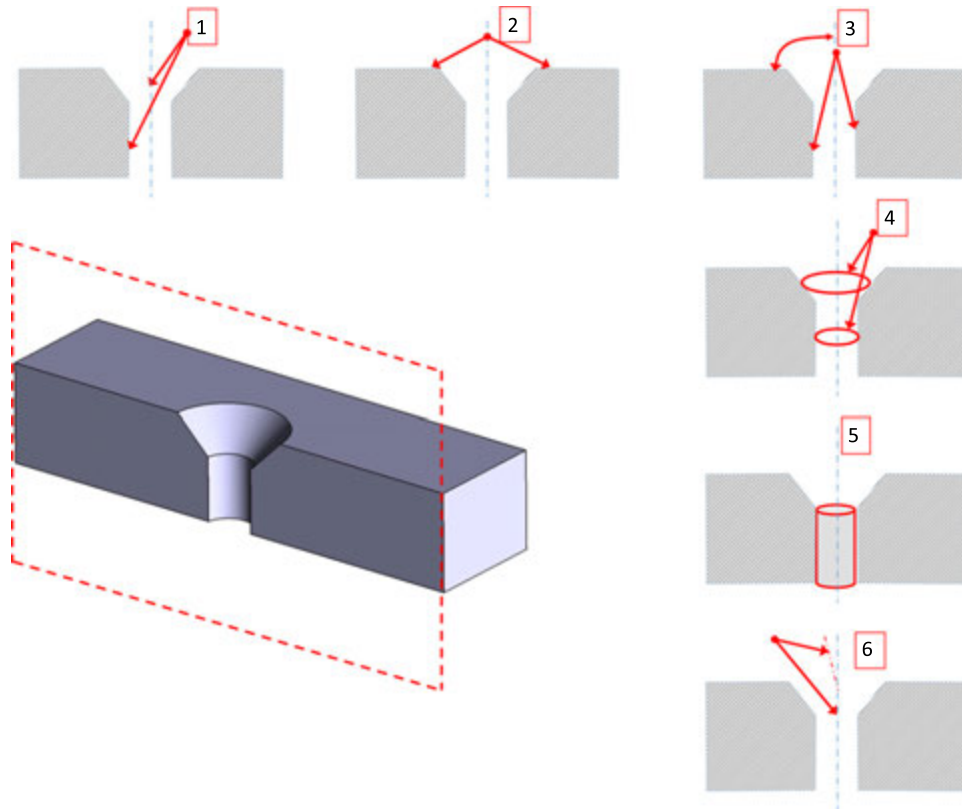


Fig. 3 Geometrical parameters for the evaluation of a countersunk hole.

components may cause a lack of contact, becoming a possible starting point to micro-cracks on the composite layer, critical in the bearing behavior of the join.

However, roughness measurements results can be affected by the position and the geometry of the holes. One of the better solutions is a novel methodology that generate better roughness measures. It consists in obtaining a polymeric replica of the drills using some replicant products that reproduce the internal surface of the hole maintaining the dimensional and the micro/macro-geometrical features. The replicant products are based on two components that solidifies into the hole without thermal effects. Once it is solidified, the material properties- in terms of elasticity, hardness and elongation- allow to use contact and non-contact measurement, ensuring an accuracy of $\pm 1 \mu\text{m}$ of reproducibility error in roughness, shape and dimensional applications.

Apart from roughness and diameter, the evaluation of drill holes can include a large number of parameters. **Fig. 3** describes the different geometrical deviations that can be detected in a countersunk hole. The six different parameters are explained as:

- (1) Straightness deviation on the axis and generatrix of the drilled hole.
- (2) Flatness deviation on the surface due to the drilling process
- (3) Parallelism deviation between the generatrix of the drilled hole. Perpendicularity between the axis of the hole and the surface of the plate.
- (4) Roundness on the drilled hole and countersink section. Concentricity between sections.
- (5) Cylindricity deviation on the hole section.
- (6) Coaxiality between hole and countersink axis.

Finally, the evaluation of the delamination is considered as one of the most relevant studies for the analysis of the drilled hole quality. Surface delamination and cracking of composite can occur in the entrance (peel-up) and exit (push-out) of the hole. The literature review about the delamination and uncut fibers inspection methods recommends the use of optical systems. Image treatment software establish the delamination area around the hole by means of addition and subtraction tools the variation of the affected area (**Fig. 4**). This process provides quantified information of the delamination, following two different equations. The delamination factor (F_d) may be calculated by the **Eq. (1)**.

$$F_d = \frac{D_{max}}{D} \quad (1)$$

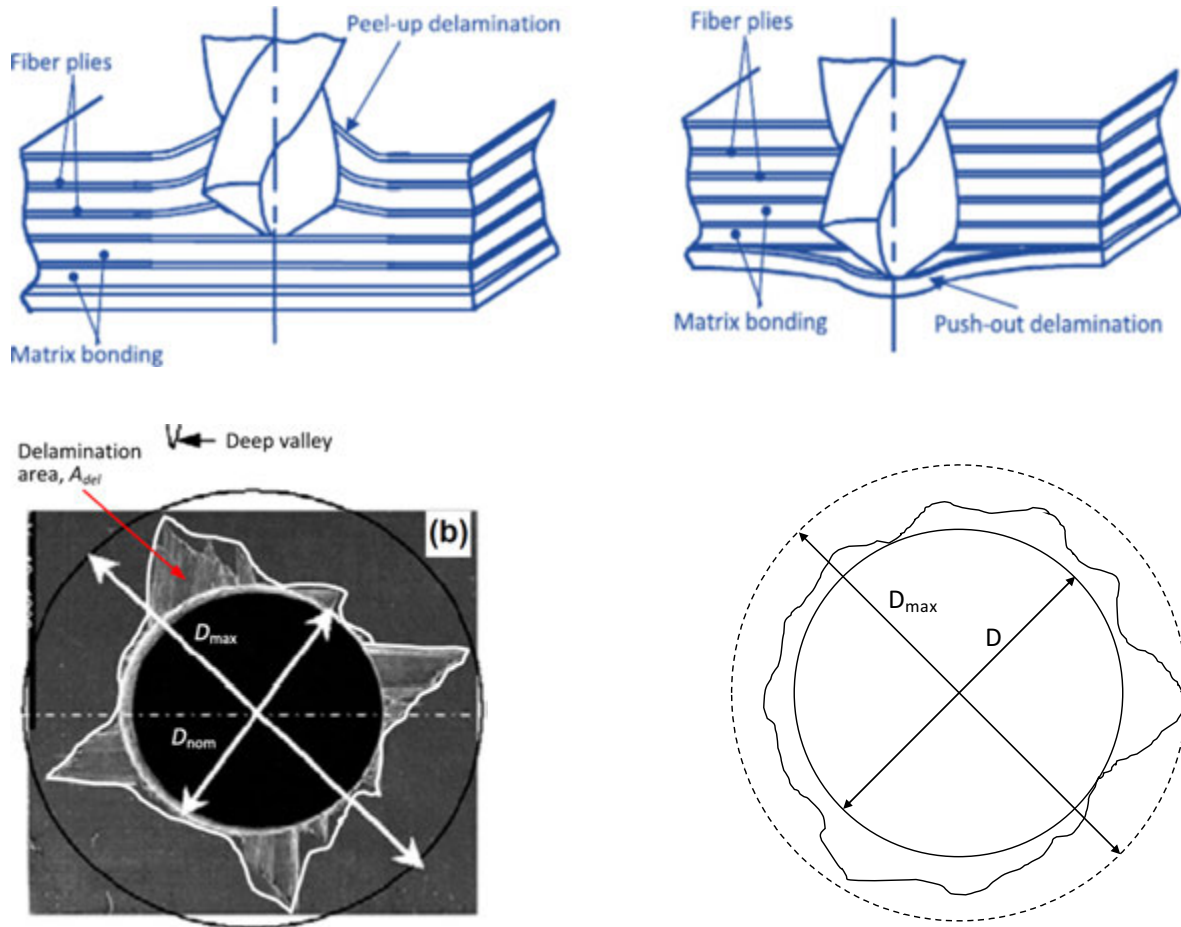


Fig. 4 Delamination effect in the drilling of CFRP.

Additionally, the delamination coefficient (DF) can be expressed as a percentage of the affected area, as shown in Eq. (2).

$$DF = \left(\frac{A_{del} - A}{A} \right) \% \quad (2)$$

Once the part is joined and in service, some dimensional measurements can be made, particularly for maintenance purposes. In this case, the instrument used is go no-go, comparator locks, thickness gauges, alexometers and internal micrometers.

This is why tool wear is not considered as a key factor and hole quality is usually over it (Dharan and Won, 2000). Additionally, another types of surface defects may be caused by thermal effects and abrasive wear phenomena on the composite hole. As shown in Fig. 5, main representative internal hole damages can be described as cracks, porosity, lack of resin between layers, etc.

Drilling Fundamentals and Operation

Drilling processes are included on the most common machining stages for composite components development, mainly due to the need of mechanical assembly parts and structures. Although it is a relatively simple cutting process, a correct understanding of the parameters involved in the manufacture of the hole is the first requirement to ensure the quality of the results.

Drilling, is a machining operation usually performed by a rotating cylindrical tool. As described in the section on this subject, hole making tools (also known as drills) generally shows two cutting edges. During cutting stage of composites, the drill and the work-piece maintains a relative movement along the hole axis. This movement are controlled by the application of thrust and torque forces adapted to drill conditions (materials, drilling equipment, tool geometry, hole quality requirements, etc.) (Groover, 2010; Teti, 2002; Davim and Technology, 2018; Abrão *et al.*, 2007). In addition, drilling forces are governed by machining parameters, being the most relevant, cutting speed and feed.

The cutting speed parameter in a drilling operation is related to the surface speed at the outside diameter of the tool. Particularly, it is determined by the spindle rotational speed and the tool features (Groover, 2010), following Eq. (3).

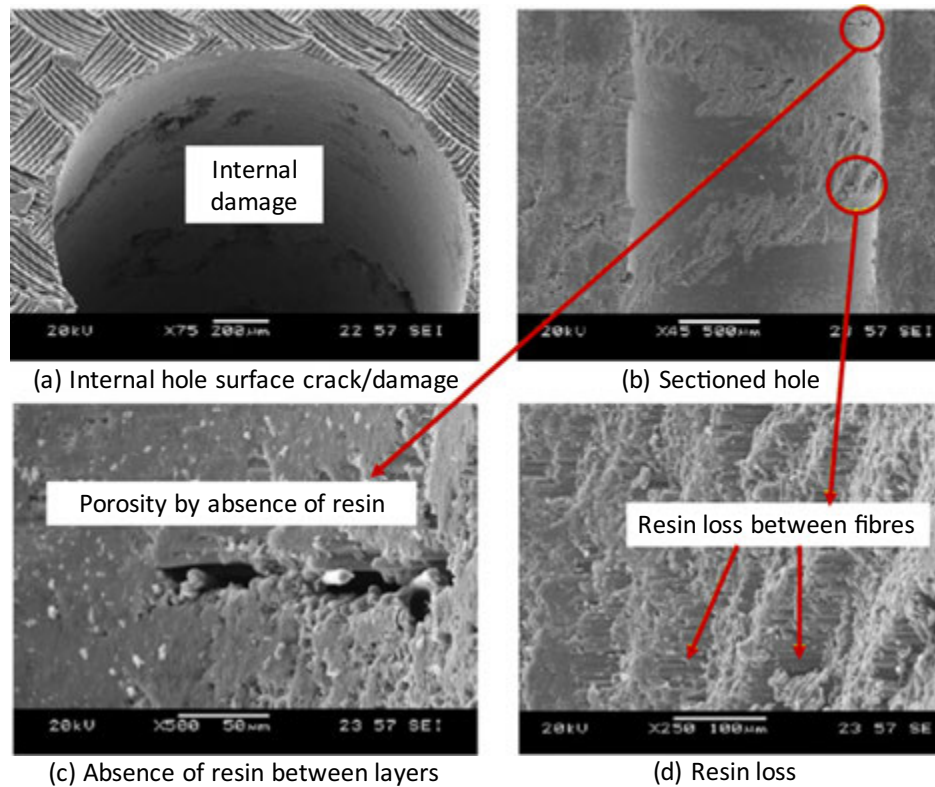


Fig. 5 Internal hole damage forms: (a) Test 11, from hole exit, hole number 250; (b) Test 3, sectioned hole, first hole; (c) absence of resin between layers (porosity) and (d) resin loss. Reproduced from Shyha, I., Soo, S.L., Aspinwall, D., Bradley, S., 2010. Effect of laminate configuration and feed rate on cutting performance when drilling holes in carbon fiber reinforced plastic composites. *J. Mater. Process. Technol.* 210, 1023–1034. doi:10.1016/j.jmatprotec.2010.02.011.

$$N = \frac{v}{\pi \cdot D} \quad (3)$$

Where N represents the spindle rotational speed [rev/min], v is the linear cutting speed [mm/min], and D quantified the drilling tool diameter in mm.

Feed (f) is one of the most important parameters in drilling operations on composite materials. It represents the axial movement of the tool as a function to the rotation of the spindle in mm/rev. Is commonly to transform to feed rate (f_r) as a function of time instead of by using the Eq. (4) (Tsao, 2012).

$$f_r = N \cdot f \quad (4)$$

Where N represents the spindle rotational speed [rev/min], f_r is the feed rate [mm/min], and f shows the feed in mm/rev.

Additionally, based on the position of the cutting tool into the composite part, some main steps can be described in the drilling process of a through hole (Davim, 2009).

In a first stage, an approximation of the drill is performed until the contact between cutting edge and work-piece. The initial contact induces a severe increase on the thrust force, and a slight increase on the torque, especially due to the small section of the tool diameter inserted into the composite material. This stage continues with a particle removing phenomenon as the cutting edge penetrates into the outer layer of the composite. This effect implies a uniform growth of the thrust and torque forces under stationary conditions, related to a complete interaction of cutting edges and work-piece. Drilling process finishes when the cutting tool comes out of the material and the cutting edges are outside the part. Under these positioning conditions, a decrease is caused in thrust and torque force due to the lack of non-machined material, as can be observed in Fig. 6.

The anisotropic and abrasive nature of composite materials as CFRP, makes that the process of drilling shows important variations from homogeneous removal applications in metallic parts. Under this consideration, the relationship between the cutting speed direction of the tool edge and the fiber composite alignment causes non-uniform forces on the edges and the increase of tool wear effects. This phenomenon provides great relevance to the composite layer design for manufacturing of components and assembly parts. Fig. 7 shows the positioning of cutting tool edges and the orientation of unidirectional fiber laminated layer in the rotational movement of the drilling process, being the main cause of the different machining forces behavior on composites (Sheikh-Ahmad, 2009; Teti, 2002; Eneyew and Ramulu, 2014; Lachaud et al., 2001).

The wide variety of the mechanical composite joining operations implies the application of different types of fasteners and head geometries and the corresponding hole (counterbored, countersunk, etc.) (Troughton, 2009). This reason makes necessary

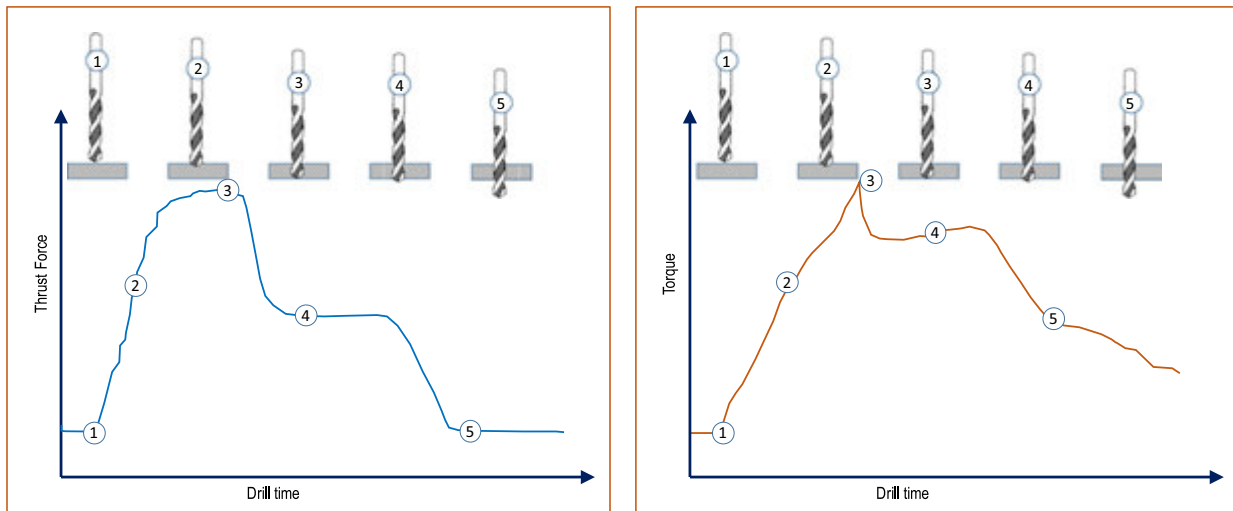


Fig. 6 Thrust and torque behavior on different stages of drilling process.

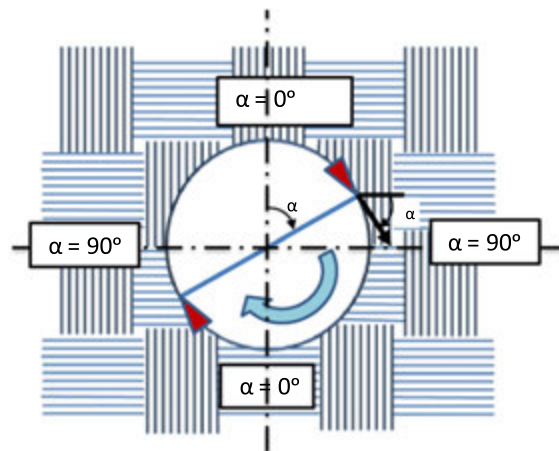


Fig. 7 Cutting edge positioning on unidirectional composite fibers.

the development of specific operations related to drilling in order to adapt the hole geometry to the surface and geometry requirements. Most of the special operations are shown as an additional operation to modify the initial properties of a hole, being necessary the development of an initial guide drill for the axis alignment of the special tools. In most of cases, the additional operation is carried out by a rotational specific tool, involving the increase of the external hole diameter size. Most common operations related to drilling are reaming, tapping, counterboring, countersinking, centering and spotfacing (Groover, 2010; Davim, 2009). Fig. 8. Shows a graphical description of these operations.

- (1) Reaming process is used to enlarge the hole diameter providing more accurate tolerance and improving the surface finish, in terms of roughness of the machined surfaces.
- (2) Tapping operation is performed to provide internal threads for screw assembly.
- (3) Counterboring processes consists in the development of greater diameter section on the drilled hole. This operation is used to avoid the protrusion of bolt heads above the surface.
- (4) Countersinking operation is very similar to counterboring. In this process the additional operation consists in the development of a cone - shaped section to seat the head of the bolts for flat surface results in assembled components.
- (5) Centering is the process to provide a starting drill for the positioning of subsequent larger diameter hole.
- (6) Spotfacing is an operation nearly to milling processes used to develop flat machined surfaces on the top of drilled holes.

The possibility of customization of composite materials combined with the possibility of optimizing their properties means that drilling processes must be continuously adapted to the market needs. This assumption makes common to find different solutions with important evolutions over time, in a process of continuous improvement that have been studied for approximately forty years.

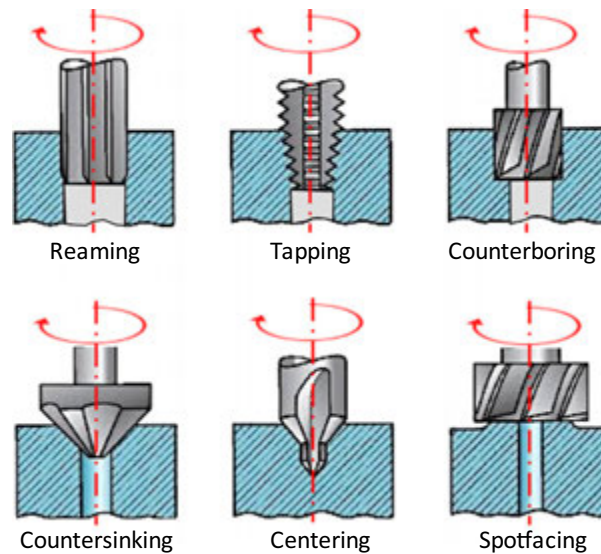


Fig. 8 Operations related to drilling processes.

Fiber Composition and Configuration Effect

The wide variety of mechanical properties that can be developed by the variation in the design based on the different special directions of the fibers (Chen *et al.*, 2019). This effect generally makes that composites show an orthotropic behavior, having dissimilar properties, strength and thermal conductivity. This fact makes more difficult the cutting stage due to lack of homogeneity of the layers and fiber directions. Therefore, the correct design of the machining process is very important to meet the established requirements. For instance, when CFRP are drilled, the edge of the cutting tool cuts the carbon fibers in different orientations by a rotation movement, while the edge moves axially along the theoretical axis of the hole. The chip produced in this case is a high abrasive short fiber dust that quickly increase tool wear, damaging the hole quality.

The composites materials design, particularly the reinforced matrix composition, is another important factor. The matrix will join the different phases of the FRP inducing special properties. They can increase the friction between the composite and the tool or reduce the temperature relief, affecting the matrix integrity and reducing the part behavior. For this reason, thermal and mechanical effects of the drilling process are usually considered in the results variation (Boccarusso *et al.*, 2019; Gaitonde *et al.*, 2008).

Summarizing, the composite design, fiber orientation and matrix composition, has an impact on the cutting process. Consequently, the proper selection of the cutting tool geometries, the machining parameters and the lubricant conditions will have a great impact on the tool wear, the final quality of the join and therefore, in the efficiency of the assembly process.

Cutting Tools

Type of Tools

Forced by the characteristics of the process and in order to reduce the effects of the fiber, cutting tool geometry for drilling FRP has been greatly improved in recent years. Their design is based on the initial design of the drill bits for metallic alloys and its continuous improvement. New geometries overcome the technological challenge of drilling CFRP, particularly when delamination issues were not solved using conventional cutting tools.

The design of new tool geometries is mostly based on empirical practice rather than in theoretical criteria. The main cutting tool geometries used for drilling CFRP are represented in Fig. 7.

Nevertheless, it should be noted that the process efficiency is obviously reduced by this method because at least two steps with different diameter drill bits should be employed.

Other relevant feature in the twist drill geometry is the chisel edge (transversal cutting lip in the point of the drill), that can contribute to the thrust force often up to 40%–60% (Won and Dharan, 2002). A way to avoid its influence is the use of pre-drilled pilot holes, with diameter equal to the diameter of the chisel edge to eliminate the extrusion effect by chisel edge (Tsao and Hocheng, 2003). This is the reason for the limited use of twist drill geometry, only when using a pilot hole still represent the best choice (Marques *et al.*, 2009).

Spur drills, is an evolution of the drill geometry used in the wood industry. They improve the surface finish keeping high productive parameters (Grilo *et al.*, 2013), but their sharpened geometry weakens the edges against abrasive wear. For this reason, this geometry is not usually seen in the drilling of CFRP.

Core drills coated with electrodeposited diamond, were often used for low thicknesses and big size diameters (>8 mm). However, the industry tends to use orbital drilling processes implemented in semi-automatic portable machines, where drilling-milling toolpaths are combined (Boccarusso *et al.*, 2019; Caggiano *et al.*, 2018).

Nowadays, the most used geometries for drilling CFRP are double point angle twist drill for small to medium size diameters (4–10 mm), dagger drills for one-shot drilling of small diameters (<5 mm) and step drills for pre-centered holes. In some cases, the design of step drills includes an edge to perform countersunk holes. They are called hybrid tools and their diameter changes produce a conical cutting at the top of the hole.

Step drills is widely used to reduce the drilling cycle time because no pre-drilling technique is needed. In this case, a drill bit diameter coefficient (k) is define to relate the primary drill bit diameter to secondary one. This coefficient has an impact in the delamination factor of hole exit, which is reduced when k is lower than 0.5 (Feito *et al.*, 2016; Qiu *et al.*, 2018a,b). It should be noticed that Step drills usually produce an entry delamination higher than exit delamination, fact that can be due to the increment in diameter (Bonnet *et al.*, 2015).

In terms of delamination, Reamers give the best results. Their straight flutes helps to avoid the effect of the machining parameters, allowing to work with high cutting parameters values and keeping a delamination factor close to one (Feito *et al.*, 2015).

Angles and Helix

Several studies of drill geometry compare new design performance with other commercial drills (Geier *et al.*, 2019). Generally, the geometry that is changed is the point angle and the helix angle, both characteristics have an impact on delamination. Low helix angles are usually used to reduce the thrust force, and consequently the delamination. At the same time, small point angle is selected for conventional twist drills. An increase in point angle extends the area involved in the extrusion effect of the uncut layers of the laminates. Therefore, a higher thrust force is obtained increasing the push-out delamination damage. This fact has also been proved for double point angle twist drills. When double point angle twist drills are selected, a point angle around 140° is recommended for the first point angle (Shyha *et al.*, 2009) and 85° (Gaitonde *et al.*, 2008; Heisel and Pfeifroth, 2012) for the second one.

Materials: Substrates and Coatings

Drills used in the machining of FRP are usually made of tungsten carbide and cobalt (WC-%Co) to improve their behavior to abrasion compared to traditional High Speed Steel drill bits. In general, the use of carbide is recommended for the better performance of the tools and a good behavior against delamination regardless of the cutting parameters used (Davim and Reis, 2003) but this type of uncoated tools have a tool life of few hundreds of holes.

Tool life is limited for the fast dulling of the cutting edge, which is produced by the brittle nature of the FRP. There is no stagnation zone for FRP drilling so the edge is constantly in contact with the abrasive fibers, speeding up the abrasive wear rate. For this reason, different material coatings can be used to increase tool life of specially designed FRP drills. They prevent thermal damage and increase the surface hardness of the tool. The range of materials covers from TiN, TiAlN / AlTiN, used for the most economical applications (Hrechuk *et al.*, 2018), to diamonds, selected for high stable operations and high serial processes (Pinho *et al.*, 2016).

On the one hand, TiN, TiAlN and AlTiN coatings improve the wear behavior at a medium cost per hole. They can reduce the delamination (Wang *et al.*, 2013) but environmental conditions, particularly temperature, may increase the oxidation of AlTiN coated layer (Wang *et al.*, 2013). This fact prevents the protection of the substrate influencing both thrust force and torque and providing similar results to uncoated tools (Fig. 7).

On the other hand, diamonds coatings are used in two different ways, as an amorphous coating of Diamond-Like Carbon (DLC) or as sintered PolyCrystalline Diamond (PCD). DLC can increase the tool life by up to 200% compared to uncoated tools (Hrechuk *et al.*, 2018) but PCD is most used coating (Geier *et al.*, 2019; Wang *et al.*, 2013). PCD coated tools offers better results against axial force, increasing the tool life 10–12, considering uncoated carbide tools as a reference (Iliescu *et al.*, 2010). Additionally, the related cutting speed can be 3 times higher when using diamond-coated tools than uncoated ones. Nonetheless, working parameters should be properly selected. The fragility of PCD make easy the appearance of fractures on the PCD layer for inadequate cutting conditions (Xu *et al.*, 2019b).

Influence of Cutting Conditions

Influence of Cutting Parameters on Delamination

In addition to the tool geometry, the cutting conditions, considering cutting speed and feed rate, also have an impact on the drilling performance. They are a key factor to achieve delamination-free composite parts by controlling the thrust force (Geng *et al.*, 2019; Romoli and Lutey, 2019). In fact, the cutting forces can separate the carbon fiber layers, favoring delamination and internal

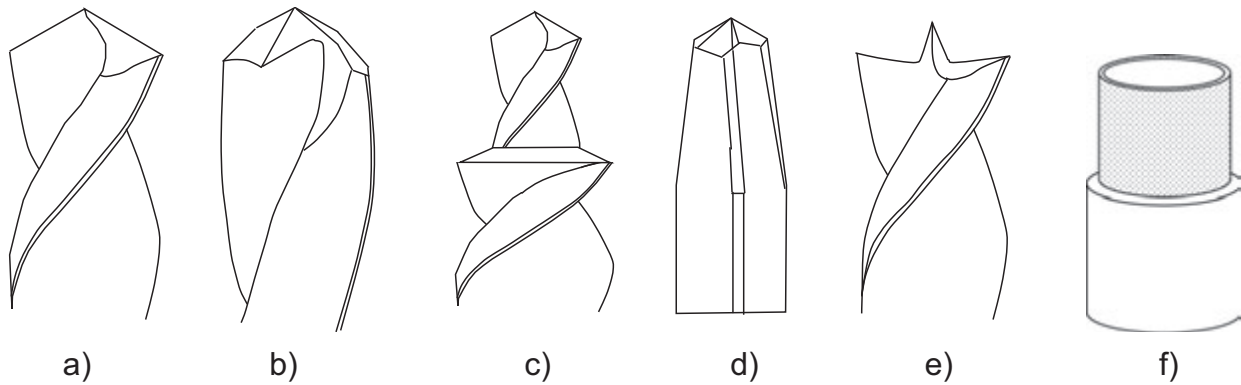


Fig. 9 Main cutting tool geometries for drilling CFRP. (a) Conventional twist drill. (b) Double point angle twist drill. (c) Step drill. (d) Reamer drill. (e) Brad & spur drill. (f) Core drill.

Table 2 Effects of drilling parameter on process behavior and hole quality

↑ Cutting speed	At higher cutting speeds, the cutting edge rubs around the hole wall more frequently, causing higher dimensional distortions (Melentiev <i>et al.</i> , 2016; Abhishek <i>et al.</i> , 2016)	↓ Thrust force ↓ Surface roughness ↑ Circularity error
↑ Feed-rate	At high feed-rates, the cutting tool cuts a higher thickness, increasing the thrust force (Melentiev <i>et al.</i> , 2016; Vinayagamoorthy, 2018)	↑ Thrust force ↑ Surface roughness ↑ Delamination
↑ Point angle	High point angles produce an increase in the contact surface between the cutting edge and the CFRP (Melentiev <i>et al.</i> , 2016; Pascual <i>et al.</i> , 2017)	↑ Thrust force ↓ Delamination
↑ Cutting diameter	As diameter increases, the contact area increases, elevating thrust force (Bonnet <i>et al.</i> , 2015; Jia <i>et al.</i> , 2016)	↑ Thrust force ↑ Delamination

defects such as fissures or cracks. For their selection, it should also be considered the infinitive options of FRP combination that can be used in the same part. This is why, it is difficult to find in the literature an optimal cutting parameter value. Nevertheless, even when cutting parameters are usually adjusted by experiments, some general lines can be followed.

As a rule, low feed-rates and high cutting speeds reduce delamination. However, critical feed-rates, which depends on the properties of the FRP, might suddenly increase them (Krishnaraj *et al.*, 2012). Some studies show through ANOVA analysis that the most influential parameter on thrust force and torque is the feed rate, linearly increasing them. The higher the feed rate, the greater the force required to penetrate the workpiece. Similarly, the minimum limit of the feed rate is defined by the receding of the individual fibers in front of the penetrating cutting edge, which decrease the quality of the hole. Despite these results, cutting speed should not be neglected due to the impact of the drill geometry of the drill on its effect (Feito *et al.*, 2016). Thrust force decreases as cutting speed increases, but up to an upper limit where the temperature achieves a maximum limit. This limit implies the risk to induce thermal damage to the composite or to soft the matrix (Rajkumar *et al.*, 2017). As a consequence, the optimum cutting speed must be high enough to allow efficient cutting of fibers with minimum heat dissipation.

Industrially, the accepted optimum speed in drilling CFRP using carbide drills is settle in ranges between 50–100 m/min. Otherwise, common feed rates values are from 0.04 to 0.2 rev/min. Slower feed rates lead to heat intensive crushing of individual fibers and increase the production time. Whereas, higher feed rates result in an increase of the axial thrust and the heat flux producing delamination.

The effect of these parameters is also related to the geometry of the tool. For example, the thrust force is more influenced by the feed rate using Brad drills than using Reamers or Step drills, as is show in Fig. 9 (Feito *et al.*, 2015). Under the same conditions, torque values for Brad drills are higher. This behavior is related to the cutting geometry and its effect on the temperature of the process. Similarly, the decrease of the thrust force related to the cutting speed is more relevant for Bar drills than to Reamers or Step drills, which generally are related to lower cutting forces. Table 2 summarize these effects and the influence of the point angle and the cutting diameter (Fig. 10).

Cooling Techniques in the Drilling of CFRP

Cooling is a useful way to reduce cutting forces and tool wear or to remove chips from the cutting area (Póka *et al.*, 2016). The main options for cooling in metal machining are conventional cutting fluid lubricants, Minimum Quantity of Lubricant (MQL) or cryogenic cooling techniques (Fig. 13).

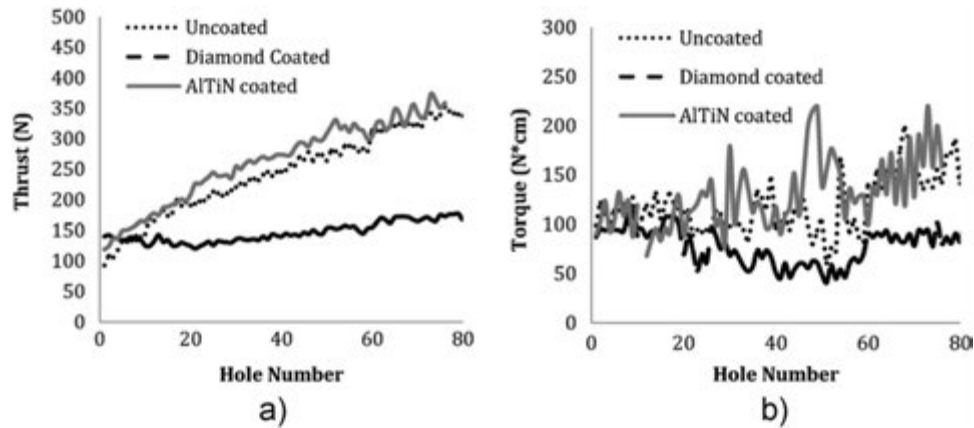


Fig. 10 Influence of tool coating in the drilling of CFRP. (a) Thrust force. (b) Torque. Reproduced from Wang, X., Kwon, P.Y., Sturtevant, C., Kim, D.D.W., Lantrip, J., 2013. Tool wear of coated drills in drilling CFRP. *J. Manuf. Process.* 15, 127–135. doi:10.1016/j.jmapro.2012.09.019.

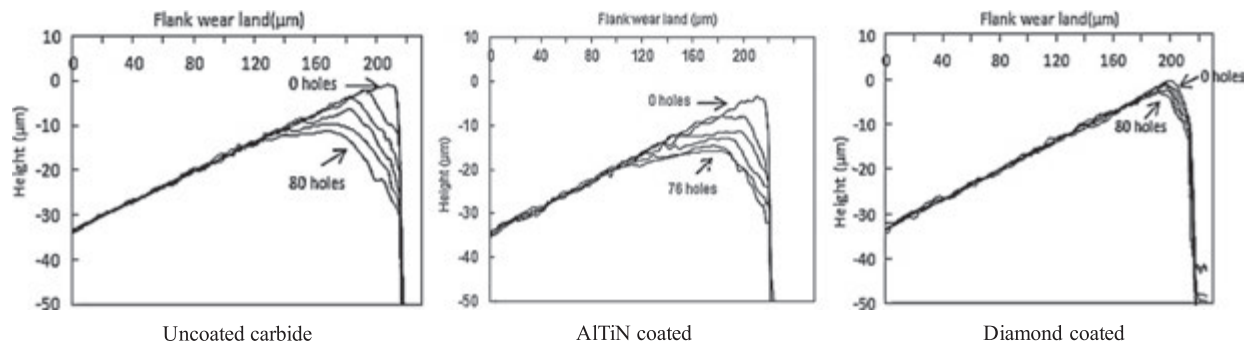


Fig. 11 Flank wear evolution on different coated CFRP drill tools. Reproduced from Wang, X., Kwon, P.Y., Sturtevant, C., Kim, D.D.W., Lantrip, J., 2013. Tool wear of coated drills in drilling CFRP. *J. Manuf. Process.* 15, 127–135. doi:10.1016/j.jmapro.2012.09.019.

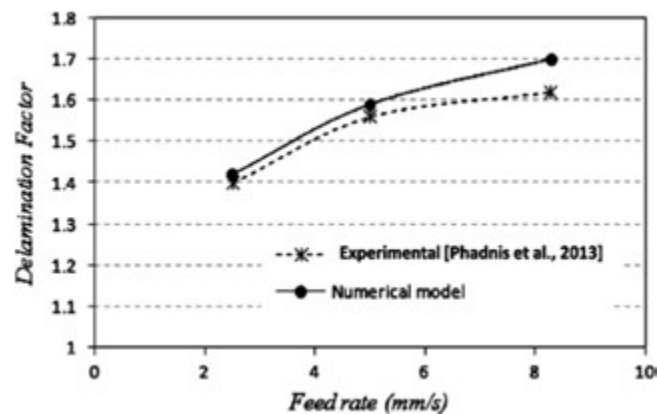


Fig. 12 Comparison of the delamination factor of numerical data with experimental data. Reproduced from Feito, N., López-Puente, J., Santiuste, C., Miguélez, M.H., 2014. Numerical prediction of delamination in CFRP drilling. *Compos. Struct.* 108, 677–683. doi:10.1016/j.compstruct.2013.10.014.

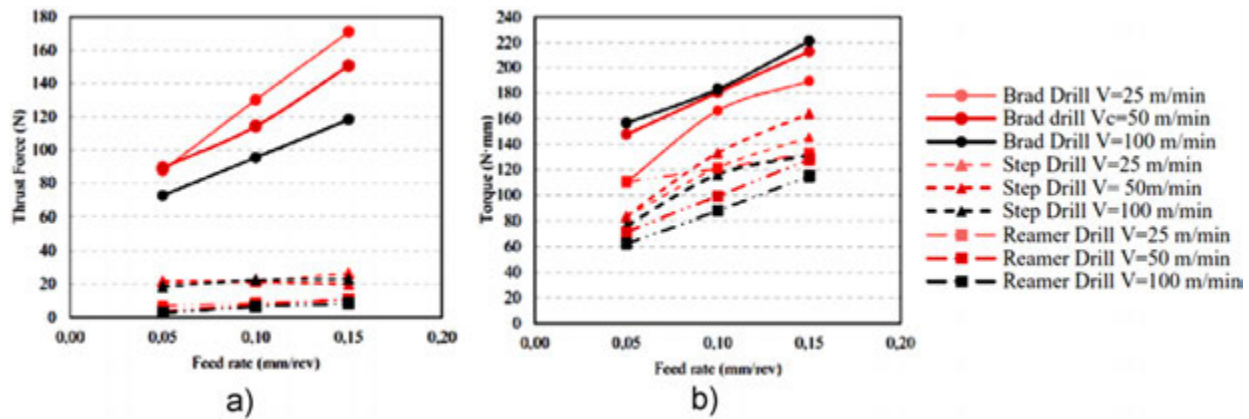


Fig. 13 Brad, Step and Reamer 6 mm drills behavior. (a) Thrust force. (b) Torque. Reproduced from Feito, N., Álvarez, J.D., Cantero, J.L., Miguélez, M.H., 2015. Influence of special tool geometry in drilling woven CFRPs materials. *Procedia Eng.* 132, 632–638. doi:10.1016/j.proeng.2015.12.541.

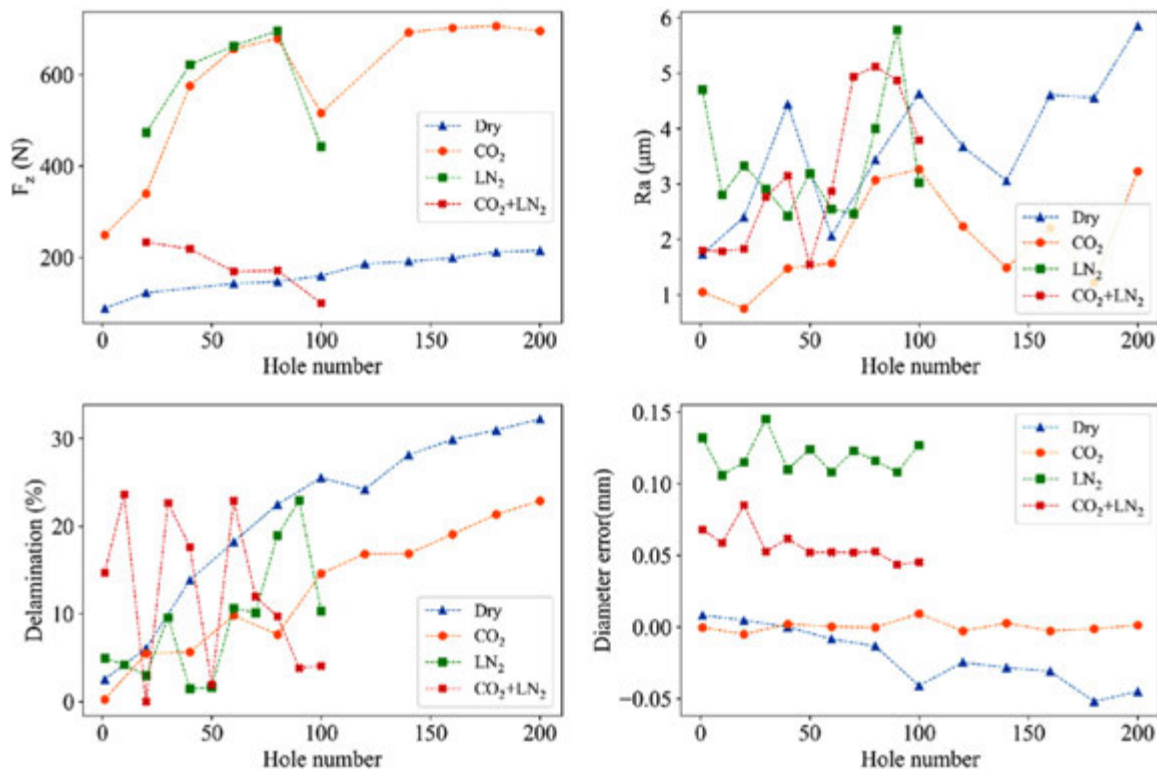


Fig. 14 Influence of cryogenic cooling on thrust force, delamination, diameter and roughness.

Conventional lubricants are not recommended for machining of FRP because carbon chips mix with the fluid lubricant creating an abrasive paste that can damage the machine guides. As a result, a vacuum cleaner is always suggested to remove carbon chips from the cutting area (Geier and Szalay, 2017).

MQL is often used as an environmentally-friendly cooling technique. It uses bio-degradable oils in a fog suspension that is conducted across the internal cooling holes of the drill bit. This small amount of lubricant does not produce an abrasive paste and it is easily retire by suction, even when it is evaporated. However, MQL does not always provide significant advantages to the process. In fact, dry drilling thrust force and torque is usually lower than MQL drilling (Kerrigan and Scaife, 2018).

Novel cooling techniques, such as cryogenic cooling using cold-air, CO₂ or LN₂, have been studied in the machining of CFRP (Morkavuk *et al.*, 2018). These techniques produce less tool damage, smaller delaminated areas and better surface roughness on the machined surfaces whereas the cutting forces increase, compared to dry milling results, due to freeze hardening. Fig. 14 shows

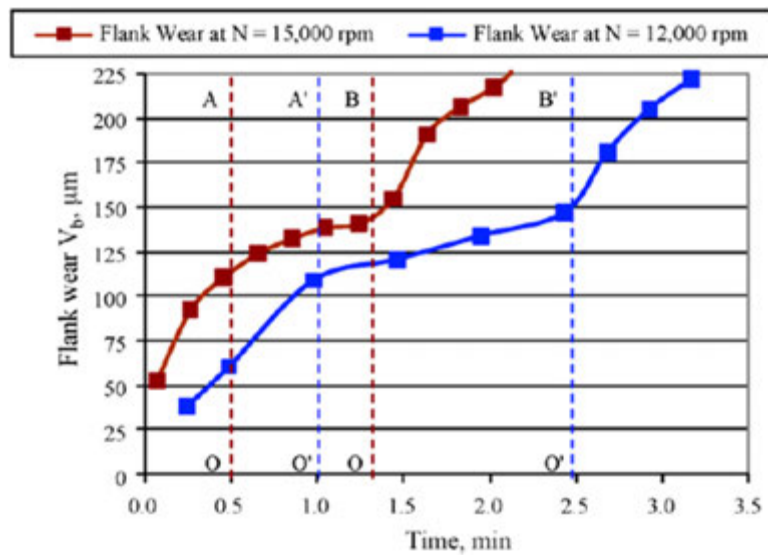


Fig. 15 Evolution of the flank wear vs. time at different cutting speed. Reproduced from Rawat, S., Attia, H., 2009. Wear mechanisms and tool life management of WC-Co drills during dry high speed drilling of woven carbon fiber composites. *Wear* 267, (5–8) 1022–1030. doi:10.1016/j.wear.2009.01.031.

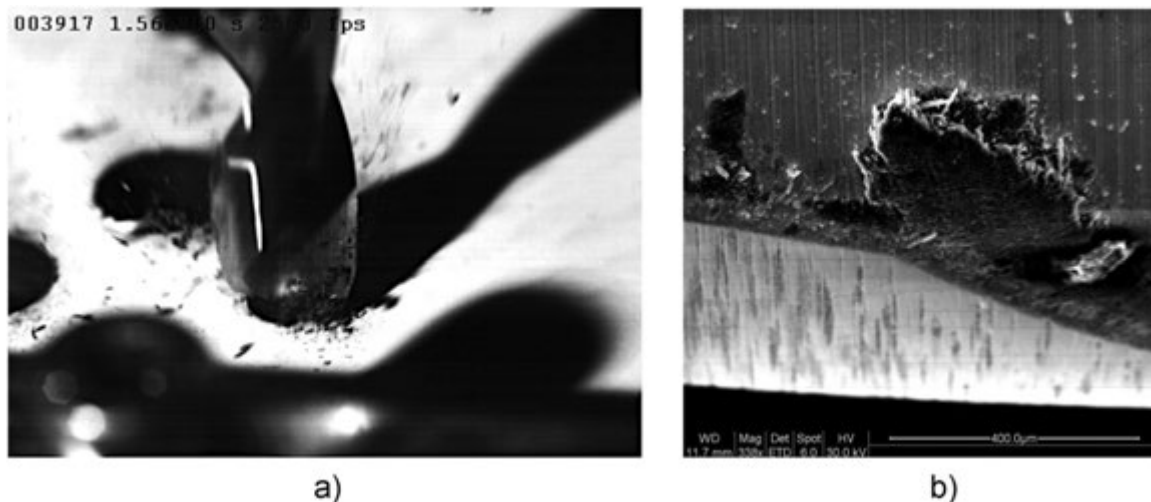


Fig. 16 (a) Composite particles detachment during drilling of CFRP (b) SEM image of tool edge showing resin adhesion combined with fibers.

the results of some drilling experiments with a dagger drill bit in four cooling conditions: dry, CO_2 throw cutting tool channels, LN_2 projected externally to the cutting tool, and the combination of $\text{CO}_2 + \text{LN}_2$.

Wear Behavior

The adverse phenomena located on the tool surface may be the cause of relevant damage effects on the composite parts such as delamination, burnt surface or geometrical deviations (Liu *et al.*, 2012; Lin and Chen, 1996; Parenti *et al.*, 2019). The apparition of defects implies an increase on the process costs, being necessary the analysis of the wear mechanisms involved in the drilling stage. As it was explained before, the composite material, depending on its composition and configuration, causes several damages to the tool as a part of the wear stage (Konig *et al.*, 1985). Moreover, its effects are related to the tool geometry, the tool substrate and coating as well as the cutting parameters used (Lin and Chen, 1996). Regarding the tool geometry, a direct relationship between wear and tool geometry where each tool shows an optimal range of use but it is also important to study the wear evolution.

Tool wear in drilling of FRP appear due the three main wear mechanisms: abrasion wear, adhesion wear and thermo-mechanical failures (Wang *et al.*, 2013; Lin and Chen, 1996). The mechanical causes, both abrasion and adhesion, follow the

traditional patterns of metal machining operations in most of the cases. Three different regions are associated to the wear phenomenon: a first phase of severe wear, a second stationary phase or controlled growth, and a third phase of life ending or severe growth (Rawat and Attia, 2009). However, for some combinations of cutting parameters, especially for high cutting speed, the evolution of the tool wear do not present a stationary wear zone, as is shown in Fig. 11.

The abrasive phenomenon is the responsible of the wear on the cutting edges. It is considered the predominant wear mechanism (Liu *et al.*, 2012; Lin and Chen, 1996; Rawat and Attia, 2009) and therefore the one that controls the wear process. The intensity of this mechanism depends on the parameter selection. Observed in most of the FRP machining, it is related to the hard and abrasive features of the reinforcement (Xu *et al.*, 2019a,b).

The critical properties changes in the matrix-reinforcement interface during machining enhancing the abrasion mechanism. Chipping and the end of the previously cut fiber, impacts into the rake face and the primary edge of the tool, producing micro-cracks on its surface (Konig *et al.*, 1985) which weaken the surface integrity of the tool (Rawat and Attia, 2009). This continuous loss of the tool material at different areas of the tool is enhanced by the fragile nature of the WC-Co tool substrate. This irregularity of the tool geometry is similar to the traditional flank wear but is accelerated due to the dynamic behavior of the chip producing delamination and geometrical deviations (Xiao *et al.*, 2017; Wang *et al.*, 2017).

Additionally, the wear effect caused by different mechanisms induces the obstruction of the tool evacuation channels, producing indirect wear. A temperature increase caused by the low thermal conductivity combined with the difficulty to the chip flow, make easy, the reinforcement to be adhered and rolled causing the fracture of the tool (Konig *et al.*, 1985). Similarly, fragments of composite materials can adhere to the tool surface due to thermal causes (Liu *et al.*, 2012; Mayuet *et al.*, 2013). The adhesion of these composite particles can also favor the abrasion when they are detached. Fig. 12 shows the defects of wear due to adhesion on the rake face and abrasion on the flank face (Figs. 15 and 16).

Similarly, cutting parameters shows high influence on the temperature of the process. In order to maximize tool life, control of wear effects by control of mechanical process parameters (force and torque) as well as temperature could lead to assurance of hole quality. Mainly due to the nature of the process with difficult to access, and without the possibility of interaction, a correct monitoring of the drilling stage is shown as a high complexity task, even more in the case of temperature measurement on the cutting area. For this reason, simulations model by finite elements (FEM) and other techniques are often used to help in the understanding of the problem. However, these simulations show great problems associated with the modeling of the different phases of the composites that makes the results obtained will be based in approximations in many cases. For example, Feito *et al.* (2014) managed to predict the delamination factor by approximating the different layers to a continuous material.

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Subject Index

Note

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Cross-reference terms in italics are general cross-references, or refer to subentry terms within the main entry (the main entry is not repeated to save space).

Location references refer to the volume number, in bold, followed by the page number.

Page numbers followed by “*f*” and “*t*” refer to figures and tables, respectively.

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